Ab initio calculation of the perpendicular giant magnetoresistance of finite Co/Cu(001) and Fe/Cr(001) superlattices with fluctuating layer thicknesses

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The results of rigorous quantum calculations of the current-perpendicular-to-plane giant magnetoresistance (CPP GMR) of finite Co/Cu(001) and Fe/Cr(001) superlattices with perfectly flat interfaces but with growth-induced fluctuations in layer thicknesses are reported. They are based on an exact numerical evaluation of the Kubo formula using tight-binding parametrization with s, p, d bands and hopping to first and second neighbors of an *ab initio* band structure. These calculations show that three distinct regimes of CPP transport occur. When there are no fluctuations, CPP transport is in the ballistic regime. The CPP GMR ratio R_{CPP} of finite Co/Cu and Fe/Cr superlattices in the ballistic regime reach saturation values equal to R_{CPP} of an infinite superlattice after only ≈ 3–5 repeats of a superlattice unit cell and the maximum values of R_{CPP} are of the order of 100%. When small fluctuations in layer thickness corresponding to only one atomic plane at the interface being displaced are introduced, transport changes from ballistic to Ohmic. The calculated GMR ratio R_{CPP} increases initially linearly with the number N of ferromagnet/spacer bilayers and then saturates for $N \approx 40$–50. The theoretical maximum values of R_{CPP} for Co/Cu and Fe/Cr superlattices in the Ohmic regime are in the region 800–1000 %. The zero-field and saturation-field resistances increase linearly with N (good Ohm's law) and the calculated zero-field resistance of the Co/Cu superlattice is within 10% of the resistance observed in a Co/Cu sample of the same composition and thickness. Small spontaneous (growth-induced) fluctuations in layer thickness can thus account well for the observed CPP GMR. When superlattices with large fluctuations in layer thickness are grown deliberately (pseudorandom spin valves), the Ohmic regime changes into, experimentally as yet unexplored, Anderson localization regime. The results for Co/Cu and Fe/Cr superlattices in which layer thicknesses are made to fluctuate typically between 2 and 10 atomic planes show that strong disorder of the sequence of ferromagnet/spacer interfaces has virtually no effect on the saturation-field resistance R_{FM}, which remains as low as in the Ohmic regime. The zero-field resistance, on the other hand, increases approximately exponentially with the number of bilayers N due to Anderson localization with a localization length ≈ 30–40 nm. The CPP GMR ratio R_{CPP}, therefore, also increases approximately exponentially with N and values as high as $R_{\mathrm{CPP}} \approx 3 \times 10^4$ are predicted for Fe/Cr valves with $N \approx 50$ bilayers. Somewhat smaller ($R_{\mathrm{CPP}} \approx 10^4$) enhancement of the CPP GMR is obtained for Co/Cu pseudorandom spin valves. The conditions under which such enhancement should be observable are discussed. [S0163-1829(97)04102-7]

I. INTRODUCTION

I recently proposed[1] that magnetic multilayers with deliberately induced large fluctuations in layer thickness should exhibit a very large enhancement of the current-perpendicular-to-plane giant magnetoresistance (CPP GMR). Such multilayers will be referred to as pseudorandom spin valves.[2] A single-orbital tight-binding model used in Ref. 1 predicts that the CPP GMR of a pseudorandom spin valve grows exponentially with its thickness and values of the GMR ratio R_{CPP} as high as $R_{\mathrm{CPP}} \approx 10^5$ % can be expected.[1] The large enhancement of the CPP GMR is due to quantum interference of electrons undergoing multiple reflections from a disordered sequence of ferromagnet/spacer interfaces. The predicted very large enhancement of the CPP GMR can be observable only if the contribution to the resistance of a pseudorandom spin valve due to quantum interference effects is so large that it dominates the total resistance of the valve. To decide whether this is the case one needs to make a rigorous quantum calculation of the total resistances in the antiferromagnetic (AF) and ferromagnetic (FM) configurations for a specific multilayer system using an *ab initio* band structure. I report here the results of such calculations based on an exact numerical evaluation of the Kubo formula for fcc Co/Cu(001) and bcc Fe/Cr(001) finite superlattices sandwiched between two semi-infinite leads. These calculations show that, depending on the size of fluctuations in layer thickness, three different regimes of CPP transport occur: ballistic, Ohmic, and Anderson localization.

The first regime corresponds to no fluctuations in layer thickness, i.e., the case of transport in a perfectly periodic but finite superlattice without any impurities. The superlattice is sandwiched between two semi-infinite leads made of the same material as the nonmagnetic spacer (Cu or Cr). When the number N_{rpt} of unit cells of such a finite superlattice is sufficiently large (typically, $N_{rpt} \approx 3$–5 is enough), the results of Schep *et al.*[3] for an infinite superlattice are recovered. This provides an independent check on the validity and accuracy of the evaluation of the GMR from the Kubo formula since Schep *et al.*[3] have used a completely different method based on counting propagating states. (Note that their method can only be applied to a perfectly periodic infinite system.) As already demonstrated by Schep *et al.*[3], a large $R_{\mathrm{CPP}} > 100\%$ due to quantum effects is obtained. However, the total resistance from this source for typical Co/Cu superlattices is only $\approx 3 \times 10^{-15}\ \Omega\ \mathrm{m}^2$, which is too small to

explain the measured results. Moreover, the calculated resistances and GMR are independent of the sample thickness which disagrees with the experiment.[4,5] One must, therefore, conclude that quantum effects in perfect periodic superlattices (ballistic transport) are not seen in the present experiments. However, they should be seen in future devices with smaller transverse dimensions (ballistic contacts[6]).

The second regime occurs for finite Co/Cu and Fe/Cr superlattices without any impurities in which the thicknesses of both the magnetic and nonmagnetic layers are allowed to deviate at random from their nominal values. The CPP GMR was again evaluated exactly from the Kubo formula. Small fluctuations in layer thickness corresponding to only one atomic plane at the interface being displaced were considered. Such small fluctuations occur spontaneously due to terrace formation even in most carefully grown superlattices. The effect of such relatively small growth imperfections on the CPP GMR is profound. The transport changes from ballistic to Ohmic and the CPP GMR increases initially linearly with the superlattice thickness and then saturates. This is precisely the behavior of the CPP GMR observed by Schroeder *et al.*[4] The calculated CPP GMR ratio R_{CPP} ranges from about 50 to 1000 % depending on the sample thickness and unit cell composition. Moreover, the total resistance in the antiferromagnetic configuration of a Co/Cu sample of the same composition and thickness as in Ref. 4, calculated without any adjustable parameters, is $\approx 165\times 10^{-15}\ \Omega\ \mathrm{m}^2$. This is almost exactly the same value as observed by Schroeder *et al.*[4]. The calculated results, therefore, indicate that the whole observed CPP GMR can be explained by quantum scattering from small fluctuations in layer thickness.

Finally, when large fluctuations in layer thickness are induced deliberately, one is in the pseudorandom spin valve regime.[1] Pseudorandom spin valves can be fabricated by growing layers whose thicknesses follow a predetermined pseudorandom sequence.[1] Once a specific pseudorandom sequence is chosen to grow an experimental sample, the CPP GMR of that particular sample can be evaluated exactly from the Kubo formula in which the same pseudorandom growth sequence is used, and vice versa. I report here the results for Co/Cu and Fe/Cr pseudorandom spin valves in which the thicknesses of the magnetic and nonmagnetic layers were typically made to fluctuate between 2 and 10 atomic planes. The calculations based on an *ab initio* band structure confirm the results obtained earlier for a single-orbital tight-binding model.[1] The calculated CPP GMR of Co/Cu and Fe/Cr pseudorandom spin valves increases approximately exponentially with the valve thickness and the maximum calculated R_{CPP} is of the order of 5×10^4 % both for Co/Cu and Fe/Cr valves.

II. FINITE PERFECTLY PERIODIC SUPERLATTICE

The only reliable quantum-mechanical method for calculating the GMR without any adjustable parameters is an exact numerical evaluation of the Kubo formula using a fully realistic band structure. In general, this is, of course, an impossible task. However, there are well defined cases for magnetic multilayers accessible to experiment for which this can be done. The simplest case is that of a perfect finite superlattice without any impurities sandwiched between two contacts (lead wires). This is one step closer to reality than the pioneering work of Schep *et al.*[3] who calculated the GMR for a perfect infinite superlattice using an *ab initio* band structure. With a finite superlattice, one has a translationally inhomogeneous system and the method of counting all the propagating states in an infinite superlattice, employed by Schep *et al.*,[3] is no longer applicable. The Kubo formula has to be used instead. It was evaluated using a tight-binding parametrization with s,p,d bands and hopping to first and second nearest neighbors of an *ab initio* band structure. The tight-binding parameters for ferromagnetic fcc Co were taken from Ref. 7 and those for Cu, Fe, and paramagnetic Cr were taken from Ref. 8. A small lattice mismatch between Co/Cu and Fe/Cr was neglected.

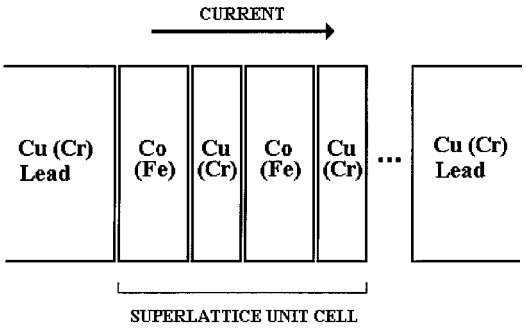

FIG. 1. Schematic representation of a finite magnetic superlattice in the CPP geometry. All the magnetic (nonmagnetic) layers have the same thickness M (N) in Sec. II but the thicknesses of both magnetic and nonmagnetic layers are allowed to fluctuate at random in Secs. III and IV.

The system for which the Kubo formula was evaluated is shown schematically in Fig. 1. It consists of the left and right semi-infinite leads (contacts) made of the spacer material (Cu or Cr) which are attached to N_{rpt} repeats of a superlattice unit cell. Each magnetic unit cell consists of a ferromagnetic layer containing M atomic planes followed by N atomic planes of a nonmagnetic spacer, a second ferromagnetic layer containing again M atomic planes and, finally, a second nonmagnetic layer of N atomic planes. For the purpose of comparing the calculated results with the experiment, the more commonly used number of bilayers N ($N=2N_{rpt}$) will be used.

The Kubo formula has to be evaluated separately for up- and down-spin carriers in the FM configuration and for carriers of either spin orientation in the AF configuration. Since the in-plane translational invariance is preserved, the wave vector $\vec{k}_{\parallel}$ parallel to the layers remains a good quantum number. It follows that the total conductance Γ^{σ} in a spin channel σ can be written for any magnetic configuration of the superlattice as a sum of partial conductances

$$\Gamma^{\sigma}=\sum_{\vec{k}_{\parallel}} (e^2/h)\Gamma^{\sigma}(\vec{k}_{\parallel}) \tag{1}$$

where $\Gamma^{\sigma}(\vec{k}_{\parallel})$ is the partial conductance in a channel $(\vec{k}_{\parallel},\sigma)$ measured in units of the quantum conductance e^2/h

and the sum in Eq. (1) is over all $\vec{k}_\parallel$ from the two-dimensional Brillouin zone (BZ). The partial conductance $\Gamma^\sigma(\vec{k}_\parallel)$ is given by the Kubo formula.[6,9–11]. For a general multiorbital band structure, the Kubo formula expressed in terms of the one-electron propagators takes the following form

$$\Gamma^\sigma(\vec{k}_\parallel)=4\mathrm{Tr}[\widetilde{G}^\sigma_{00}(\vec{k}_\parallel)t_{01}(\vec{k}_\parallel)\widetilde{G}^\sigma_{11}(\vec{k}_\parallel)t_{10}(\vec{k}_\parallel) - \mathrm{Re}(\widetilde{G}^\sigma_{01}(\vec{k}_\parallel)t_{10}(\vec{k}_\parallel)\widetilde{G}^\sigma_{01}(\vec{k}_\parallel)t_{10}))]. \quad (2)$$

Equation (2) is a straightforward generalization of the result obtained earlier by Lee and Fisher[10] (see also Ref. 11) for a single-orbital tight-binding model. The indices 0, 1 in Eq. (2) label any two neighboring principal planes[12] parallel to the layer structure, $\widetilde{G}_{i,j}(\vec{k}_\parallel)=(1/2i)[G^-_{i,j}(\vec{k}_\parallel)-G^+_{i,j}(\vec{k}_\parallel)]$, 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, $t_{01}(\vec{k}_\parallel)$ 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. The trace is taken over all the orbital indices that are contained implicitly in the principal layer indices 0,1. Since hopping to nearest and second nearest neighbors is considered, each principal plane contains two atomic (001) planes and, therefore, all the Green's functions and hopping matrices in Eq. (2) are 18×18 matrices. Finally, the usual GMR ratio, defined in terms of the conductances for the ferromagnetic (FM) and antiferromagnetic (AF) configurations of the magnetic layers, is given by

$$R_{\mathrm{CPP}}=(\Gamma^\uparrow_{\mathrm{FM}}+\Gamma^\downarrow_{\mathrm{FM}}-2\Gamma^{\uparrow,\downarrow}_{\mathrm{AF}})/2\Gamma^{\uparrow,\downarrow}_{\mathrm{AF}}. \quad (3)$$

The input in Eq. (2) are the matrix elements of the one-electron Green's function in and between the principal planes 0, 1. This is exactly the same information that is needed in the calculation of the oscillatory exchange coupling from the spin-current formula.[13,14] The formal similarity between the spin current formula for the coupling[13] and the Kubo formula for CPP transport merely reflects the fact that the two effects are closely related. The oscillatory coupling is determined by the spin current between the magnetic layers and the CPP GMR by the electric current but both currents are, of course, carried by the same electrons. This observation alone is a compelling reason for making a fully quantum calculation of the CPP GMR. Without quantum interference effects, there would be no coupling and it is, therefore, most likely that the quantum effects are very important also in the CPP transport.

From the technical point of view, the formalism for calculating local one-electron Green's functions has already been developed for oscillatory exchange coupling[14] and it can be easily adapted to the present problem. One uses the trick of cutting formally the multilayer between the planes 0 and 1 into two disconnected parts by setting the hopping matrix h_{01} equal to zero. The two disconnected parts are referred to as the left and right overlayers on semi-infinite leads.[1] It is convenient to make the cut between the finite superlattice and the left semi-infinite lead. The next step is the calculation of the surface Green's functions for the left and right overlayers. I use notation g^σ_{00} for the left and g^σ_{11} for the right overlayer surface Green's functions. Finally, the exact Green's functions G^σ_{00}, G^σ_{11}, and G^σ_{01} for the connected multilayer are obtained from g^σ_{00} and g^σ_{11} using the Dyson equation.[1]

The Green's function g^σ_{00} is simply the surface Green's function of the left semi-infinite lead (Cu or Cr). The surface Green's function g^σ_{11} of the right overlayer is generated recursively[10,15] from the surface Green's function of the right semi-infinite lead. All the atomic planes of the right overlayer are deposited one by one on the right lead and the overlayer surface Green's function is updated after each deposition from the Dyson equation:

$$[g^\sigma_{\mathrm{new}}(\vec{k}_\parallel)]^{-1}=[g^\sigma_{\mathrm{isol}}(\vec{k}_\parallel)]^{-1}-t_{01}g^\sigma_{\mathrm{old}}(\vec{k}_\parallel)t_{10}, \quad (4)$$

where $g^\sigma_{\mathrm{isol}}(\vec{k}_\parallel)$ is the Green's function of an isolated principal layer of the material that is being deposited. Provided the surface Green's function of the lead is known, the recursive method based on repeated application of Eq. (4) (method of adlayers[15]) involves no approximations and, therefore, gives the Green's functions of the connected multilayer with a machine accuracy. Moreover, the technique is not restricted to a periodic system and this flexibility of the method of adlayers will be exploited to the full in Secs. III and IV.

The only remaining problem is, therefore, the calculation of the surface Green's functions of the left and right semi-infinite leads. In our previous calculations[14] of the exchange coupling in Co/Cu(001), we used an iterative decimation technique.[16] In this method, the surface Green's function is approximated by its value at the surface of a thick stack of atomic planes. However, to obtain a truly surface Green's function, it is necessary to add in the decimation method a small imaginary part ϵ to the energy to disrupt quantum interference between the two surfaces of the slab. When ϵ is small, the convergence of the decimation method becomes poor. This is not a problem in total energy calculations since one integrates over a contour in the complex energy plane. However, there is no energy integral in the Kubo formula (transport takes place at the Fermi surface) and ϵ has to be very small in order not to introduce a spurious resistance due to finite lifetime effects. I have, therefore, used an entirely new noniterative technique for generating the surface Green's function[17] in which the convergence problem does not arise. A value $\epsilon=10^{-8}$ Ry, which was used in all the calculations, is so small that it has no effect on the conductance.

Finally, the BZ sum in Eq. (1) over $\vec{k}_\parallel$ needs to be carried out. The convergence in $\vec{k}_\parallel$ is not such a serious problem as in the calculation of the oscillatory exchange coupling. This is because, unlike the coupling, the GMR effect does not decrease with increasing thickness of the multilayer. (I am not interested here in details of quantum oscillations of the GMR about its average value[18,19] which would require a far higher accuracy.) Nevertheless, a large number of $\vec{k}_\parallel$ points in the two-dimensional (2D) BZ is needed to determine even the nonoscillatory part of the GMR. In all the calculations reported here, I first used $10^4\vec{k}_\parallel$ points in the 2D BZ and then checked the result for convergence with 4×10^4 points. This number of $\vec{k}_\parallel$ points is sufficient to achieve convergence for

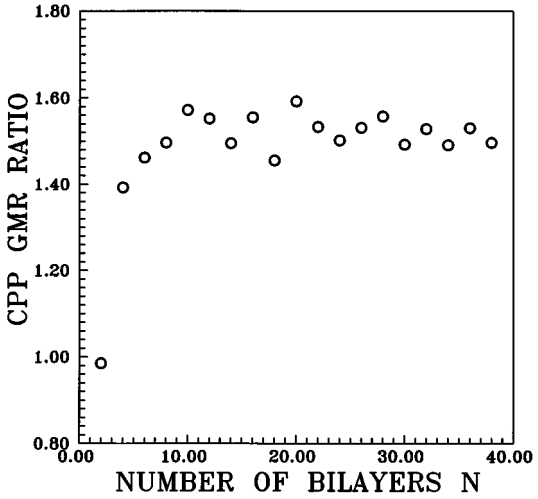

FIG. 2. Dependence of the CPP GMR ratio of a finite Co_5Cu_5 superlattice on the number of bilayers N.

multilayers of up to 150 nm thick. This means that, for superlattices with about fifteen atomic planes per unit cell, the total number of bilayers that can be handled is $N \approx 100$. For larger unit cells, reliable results can be obtained only for $N \leqslant 50$.

The calculated CPP GMR ratio for Co_5Cu_5 superlattice is plotted in Fig. 2 against the number of bilayers N. The results for Co/Cu superlattices with other compositions of the unit cell and those for Fe/Cr superlattices are qualitatively the same. A typical feature is that after only about three repeats of the unit cell the CPP GMR ratio R_{CPP} for all the superlattices investigated reaches a saturation value equal to R_{CPP} of an infinite superlattice. Oscillations about the saturation value seen in Fig. 2 are a genuine effect and occur due to size quantization in a superlattice of a finite thickness.

Given that the average nonoscillatory component of the CPP GMR reaches its saturation value so rapidly, there is no need to discuss here finite superlattices in any great detail since the results of Schep *et al.*[3] for infinite superlattices are already a very good guide to their behavior. In the case of Co_5Cu_5 superlattice, the saturation value of GMR is $R_{\mathrm{CPP}} \approx 150$ %, which is very close to the result obtained by Schep *et al.* (120%) for an infinite Co/Cu superlattice of the same composition. The small discrepancy between the present results and those of Schep *et al.*[3] is due to differences in the band structures used. (Schep *et al.*[3] considered a tetrahedral distortion of the Co lattice to allow for a small lattice mismatch between Co and Cu which is neglected in the present work.) When the conductance calculated from the Kubo formula for pure Cu is compared with the conductance obtained by counting the propagating states, the results are identical[19] and equal to the value of the ballistic conductance of pure Cu quoted by Schep *et al.*[3,6]

The fact that the calculated CPP GMR is almost as large as the observed effect[4] is not sufficient. It is also necessary to check whether the total resistance of the sample, particularly in its AF configuration, is large enough to be measurable. The results of the present calculation, in agreement with Schep *et al.*,[3] give $2\Gamma_{\mathrm{AF}} \approx 3 \times 10^{14}\ \Omega^{-1}\ \mathrm{m}^{-2}$. It follows that the ballistic resistance R_{AF}, which is independent of the superlattice length, is only about $3 \times 10^{-15}\ \Omega\ \mathrm{m}^2$. On the other hand, the observed[4] total resistance in the AF figuration can be as large as $175 \times 10^{-15}\ \Omega\ \mathrm{m}^2$. Moreover, the observed resistance increases linearly with increasing thickness of the multilayer whereas the ballistic resistance is independent of the thickness. This indicates that purely ballistic effects are not seen in the present experiments. However, it would be quite wrong to conclude that quantum reflections from perfectly flat interfaces play no significant role in CPP transport without first investigating the effect of small fluctuations in layer thickness which inevitably occur even in most carefully grown superlattices.

III. FINITE SUPERLATTICE WITH SMALL FLUCTUATIONS IN LAYER THICKNESS

In Sec. II perfect finite superlattices were investigated. Real samples contain imperfections. Even if the impurity concentration is negligible (e.g., for samples smaller than the mean free path), there are always growth imperfections in any layer structure. They arise because the control over layer thicknesses in deposition cannot be perfect and also because of spontaneous terrace formation. The method of adlayers combined with the Kubo formula allows us to determine exactly the CPP GMR and the individual conductances in the FM and AF configurations for multilayers in which layer thicknesses deviate at random from their nominal values. I will make the most optimistic assumption that individual layer thicknesses in experimental samples are controlled so well that they fluctuate at random by no more than one atomic plane. This restriction will be relaxed in Sec. IV.

The method for calculating the conductances and GMR ratio from Eqs. (1)–(3) is exactly the same as for a periodic superlattice but the layer thicknesses used in the adlayering procedure (4) are now selected according to the following prescription. Pseudorandom sequences $\{M_i\}$ and $\{N_i\}$ of integers distributed uniformly over intervals $[M_{\min}, M_{\max}]$ and $[N_{\min}, N_{\max}]$ are generated and the thicknesses M_i and N_i of the magnetic and nonmagnetic layers (measured in numbers of atomic planes) are chosen to follow these sequences. Since fluctuations of only one atomic plane are allowed, the conditions $M_{\max} - M_{\min} = 1$ and $N_{\max} - N_{\min} = 1$ are imposed.

To study systematically the conductances and CPP GMR of Co/Cu samples, the nominal thickness of Cu spacer was fixed between $N_{\min} = 5$ and $N_{\max} = 6$ and nominal Co thicknesses were in the range $2 \leqslant M_{\min} \leqslant 8$ ($3 \leqslant M_{\max} \leqslant 9$). The choice of the Cu thickness is dictated by the fact that the coupling should be antiferromagnetic. The thickness of Co layers was restricted to relatively small values to keep the computer time within reasonable limits. The calculated CPP GMR for a $Co_{8-9}Cu_{5-6}$ superlattice is plotted as a function of the number of bilayers N in Fig. 3 for $2 \leqslant N \leqslant 50$. The superlattice with this particular composition of the unit cell was selected for Fig. 3 because Co/Cu(111) samples with the same Co and Cu layer thicknesses were investigated by Schroeder *et al.*[4]

It can be seen from Fig. 3 that small fluctuations in layer thickness have a profound effect on the GMR. The magnitude of the calculated GMR ratio R_{CPP} is now so large that it can easily account for the whole observed effect (this is not the case for the purely ballistic contribution discussed in Sec. II). Moreover, the transport is clearly no longer ballistic and

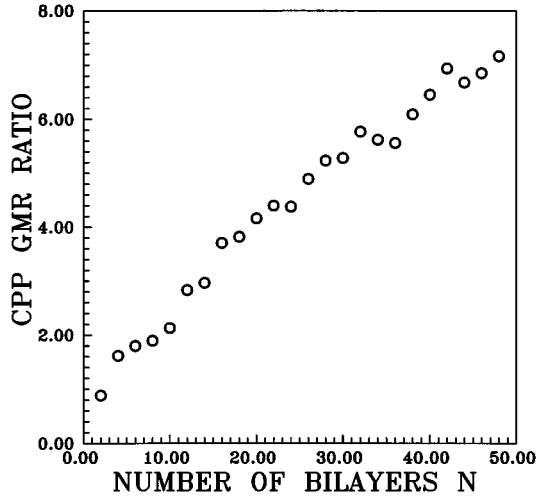

FIG. 3. CPP GMR ratio of a $Co_{8-9}Cu_{5-6}$ superlattice with small fluctuations in layer thickness plotted against the number of bilayers N.

the calculated CPP GMR appears to increase linearly with N. However, the linear dependence of the CPP GMR on the number of bilayers N holds only for relatively small values of N. For larger N, the CPP GMR reaches a saturation value. This cannot be seen in Fig. 3 because, for computational reasons, the maximum N is limited to $N \leqslant 50$. However, for a $Co_{2-3}Cu_{5-6}$ superlattice, fully converged results can be obtained for $N \leqslant 100$. They are shown in Fig. 4 (open circles) together with the corresponding results for $Fe_{2-3}Cr_{5-6}$ superlattice (full circles). It can be seen that the GMR ratio R_{CPP} reaches a saturation value of the order of 800–1000 % for both the Co/Cu and Fe/Cr superlattices. This is precisely the behavior reported by Schroeder *et al.*[4] for their Co/Cu samples. The observed initial increase of the CPP GMR followed by a saturation is due to two factors: (i) the measured resistances R_{FM} and R_{AF} in the FM and AF configurations obey Ohm's law; (ii) the values of R_{FM} and R_{AF} extrapolated to $N=0$ are very nearly equal to one another.

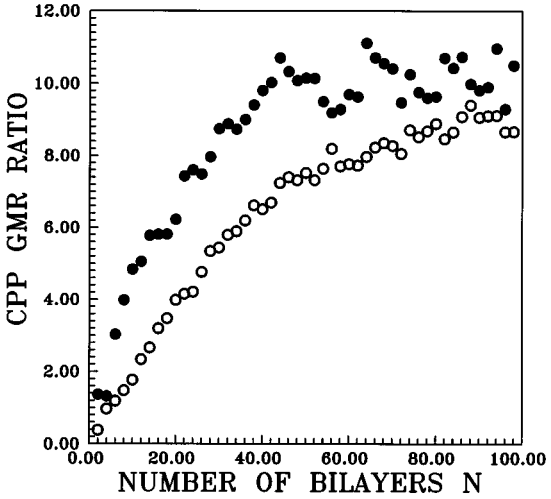

FIG. 4. CPP GMR ratios of $Co_{2-3}Cu_{5-6}$ (open circles) and $Fe_{2-3}Cr_{5-6}$ (full circles) superlattices with small fluctuations in layer thickness plotted against the number of bilayers N.

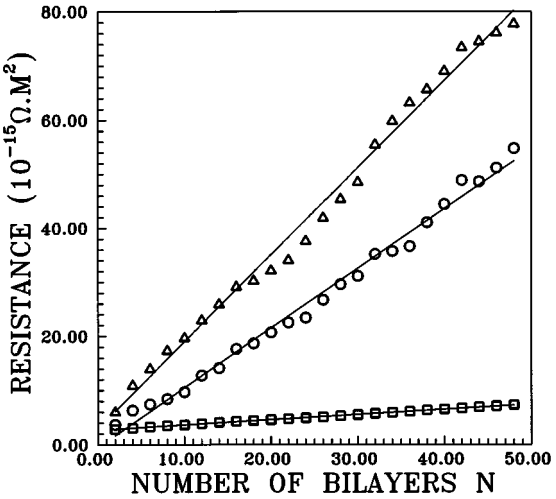

FIG. 5. Resistances in the ferromagnetic (FM) and antiferromagnetic (AF) configurations of a $Co_{8-9}Cu_{5-6}$ superlattice with small fluctuations in layer thickness plotted against the number of bilayers N. Circles denote the resistance $R_{AF}^{\uparrow,\downarrow}$ squares the resistance $R_{FM}^{\uparrow}$; and triangles the resistance $R_{FM}^{\downarrow}$.

The crucial test of the theory is, therefore, whether the absolute values of the resistances R_{FM} and R_{AF} calculated without any adjustable parameters possess these two properties. Moreover, they must also be of the same order of magnitude as the measured resistances. The theory has one advantage over the experiment in that the resistances $R_{FM}^{\uparrow}$ and $R_{FM}^{\downarrow}$ in the up- and down-spin channels in the FM configuration can be calculated separately. They are plotted in Fig. 5 together with the resistance $R_{AF}=(1/2)R_{AF}^{\uparrow,\downarrow}$ in the AF configuration against the number of bilayers N for the $Co_{8-9}Cu_{5-6}$ superlattice.

It can be seen from Fig. 5 that the calculated resistances in all three channels obey an almost perfect Ohm's law and start from approximately the same value for small N. A linear dependence on N clearly indicates that the calculated CPP resistances are due mainly to scattering from interfaces. In fact, an alternative way of viewing a superlattice with small fluctuations in layer thickness is to regard such a system as a perfectly periodic superlattice in which single atomic planes of a wrong type (Cu instead of Co, and vice versa) are inserted at random at the interfaces. The calculated CPP resistances can be then explained as being due to scattering from ''impurity'' planes located at the interfaces. The linearity of the effect indicates that the scattering from different interfaces is uncorrelated in this regime.

The calculated zero-field resistance R_{AF} of the $Co_{8-9}Cu_{5-6}$ superlattice (circles in Fig. 5) can be compared with the experimental results of Schroeder *et al.*[4] It is only necessary to extrapolate linearly the calculated R_{AF} from $N=50$ to $N=150$, which is the thickness of the sample investigated in Ref. 4. The theoretical resistance for $N=150$ is $R_{AF}=165\times10^{-15}\ \Omega\ m^2$. This is almost exactly the same value as the resistance $R_{AF}=175\times10^{-15}\ \Omega\ m^2$ measured by Schroeder *et al.*[4] for a Co/Cu(111) superlattice of the same composition and thickness ($N=150$). The results for the $Co_{2-3}Cu_{5-6}$ and $Fe_{2-3}Cr_{5-6}$ superlattices, shown in Fig. 6, are very similar and demonstrate that a good Ohm's law holds also for larger N, which provides a justification for the

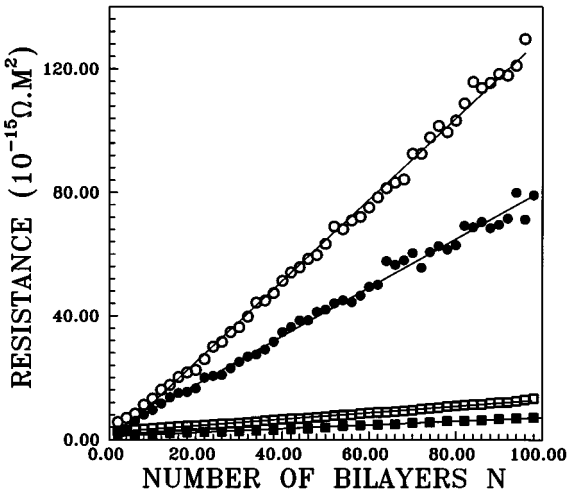

FIG. 6. Total resistances R_{AF} and R_{FM} in the antiferromagnetic (AF) and ferromagnetic (FM) configurations of $Co_{2-3}Cu_{5-6}$ (open symbols) and $Fe_{2-3}Cr_{5-6}$ (full symbols) superlattices with small fluctuations in layer thickness plotted against the number of bilayers N. Circles denote the resistance R_{AF} and squares the resistance R_{FM}.

linear extrapolation used above. Only the total resistances R_{FM} (squares) and R_{AF} (circles) in the FM and AF configurations are shown in Fig. 6. Qualitatively the same behavior is obtained for all the Co/Cu and Fe/Cr superlattices investigated with nominal Co (Fe) thickness ranging from two to nine atomic planes and nominal Cu (Cr) thicknesses fixed between 5 and 6 atomic planes.

The behavior of the resistance $R_{FM}^{\uparrow}$ in the up-spin channel in the FM configuration of the Co/Cu superlattice (squares in Figs. 5 and 6) is also very interesting. Because the matching of the up-spin bands in Co to the Cu bands is almost perfect, there is virtually no scattering at the interfaces and, therefore, $R_{FM}^{\uparrow}$ increases only very slowly with N. Up-spin (majority) electrons thus provide a low-resistance channel which shunts the high-resistance channel $R_{FM}^{\downarrow}$ (triangles in Fig. 5). It follows that, for all practical purposes, $R_{FM}^{\uparrow}$ denoted by squares in Fig. 5 can be regarded as the total saturation field resistance R_{FM}. The slow increase of R_{FM} with N is again very similar to the observed behavior.[4] However, the calculated values of R_{FM} are a factor of 3 smaller than the observed results. This is the main reason why the theoretical CPP GMR ratio is also higher by approximately the same factor than the observed R_{CPP}. One can think of two most likely explanations for this discrepancy. The first one is that there is some additional weak spin-independent scattering in the experimental samples that is not included in the present calculation. The background resistance due to such scattering masks the intrinsic scattering from Co/Cu interfaces in the up-spin channel and determines the observed $R_{FM}^{\uparrow}$. Alternatively, the matching of the Co and Cu bands in the up-spin channel may not be so perfect when one allows for relaxation effects due to a small Co/Cu lattice mismatch. On the other hand, the resistances in the down-spin channel and in the AF configuration are clearly totally dominated by the intrinsic spin-dependent scattering from Co/Cu interfaces and any background scattering (if present) is unimportant. Exactly the same arguments apply to the Fe/Cr superlattice with the only modification that the roles of the up- and down-spin channels are interchanged. This is due to the fact that it is now the down-spin (minority) band in Fe that matches almost perfectly the bands of Cr.

Before I leave this section, a comment on the nature of randomness in samples with small fluctuations in layer thickness is called for. I used a pseudorandom number sequence to model such fluctuations. One could argue that a configuration average is required. However, when different pseudorandom sequences are tried, one finds that the calculated conductances are insensitive to the choice of the sequence. This might seem surprising to those familiar with a large body of theoretical work on one-dimensional disordered wires. However, the explanation is simple. One must remember that the multilayer is not a strictly one-dimensional system. For any fixed configuration of interfaces, electrons in different $\vec{k}_{\parallel}$ channels sample different pseudorandom potential reliefs. Since the total conductance given by Eq. (1) is the sum over all $\vec{k}_{\parallel}$, some averaging over disorder is built in naturally.

IV. CO/CU(001) AND FE/CR(001) PSEUDORANDOM SPIN VALVES

Having established in Sec. III that quantum reflections from perfectly flat interfaces in multilayers with small fluctuations in layer thickness lead to CPP resistances that not only display the observed Ohmic behavior but also have the correct magnitude, one can address with some confidence the interesting questions concerning transport in pseudorandom spin valves with deliberately induced large fluctuations in layer thickness.[1]

The results of Sec. II show that the CPP GMR ratio R_{CPP} for a superlattice in the ballistic regime is independent of its thickness (for $N_{rpt} \geqslant 3-5$) and the maximum attainable R_{CPP} is only of the order of 100 %. The GMR ratio R_{CPP} of a superlattice in the Ohmic regime also saturates as a function of the number of bilayers N and the maximum theoretical attainable value of R_{CPP} for Co/Cu and Fe/Cr superlattices is of the order of 1000 %. Saturation of R_{CPP} is a consequence of the Ohmic behavior of the resistances in all three conductance channels ($R_{FM}^{\uparrow}$, $R_{FM}^{\downarrow}$, R_{AF}) and is, therefore, inevitable in this regime. The saturation value of R_{CPP} is determined by the magnetic contrast of an individual interface. 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, and Co/Cu or Fe/Cr are probably the best combinations, this places an upper bound on what can be achieved with conventional periodic superlattices in the ballistic and Ohmic regimes.

Since the CPP GMR ratio (3) expressed in terms of the resistances in the FM and AF configurations has the form

$$R_{CPP}=\frac{R_{AF}-R_{FM}}{R_{FM}} \tag{5}$$

it is clear that the only way to enhance the CPP GMR ratio beyond its saturation value is to fabricate a magnetic multilayer that operates in a regime in which the dependence of the resistance on the number of bilayers N is nonlinear. To

achieve this goal, I proposed[1] that one should grow superlattices with deliberately induced large fluctuations in layer thickness. Since CPP transport in a multilayer takes place in independent $\vec{k}_{\parallel}$ channels, the whole multilayer can be regarded as a system of one-dimensional wires connected in parallel. Large fluctuations in layer thickness mean[1] that electrons in every channel move in a one-dimensional strongly disordered potential. It follows that Anderson localization[20] must set in in every $\vec{k}_{\parallel}$ channel provided the number of bilayers N is large enough. Since the resistance in each channel increases in the localization regime exponentially with the number of bilayers N, the condition that the resistances R_{FM} and R_{AF} in the FM and AF configuration should be nonlinear functions of N can be satisfied.

It is well known[20] that the localization length decreases with increasing degree of disorder. The crucial point for magnetic multilayers is[1] that the degrees of disorder seen by electrons in the FM and AF configuration and, hence, the corresponding localization lengths are very different. Moreover, since a strong enough applied magnetic field can effect transition from the AF to the FM configuration, the degree of disorder can be controlled by the applied field. In fact, we have shown in Sec. III that matching of the Co up-spin band to the Cu bands is almost perfect. It follows that, regardless of the size of fluctuations in layer thickness, up-spin electrons in the FM configuration are only weakly scattered. One can, therefore, expect that localization either does not occur in this channel at all or is extremely weak (all the localization lengths for up-spin electrons are long). On the other hand, electrons in the down-spin channel in the FM configuration and electrons of either spin orientation in the AF configuration experience highly disordered potentials and should undergo strong localization (localization lengths in all these channels should be short).

Exactly the same arguments apply to Fe/Cr pseudorandom spin valves with the only modification that the roles of the up- and down-spin channels in the FM configuration are interchanged. In either case, the channel with a weak localization in the FM configuration shorts the channel with a strong localization and, therefore, R_{FM} should increase only slowly with the number of bilayers N. On the other hand, R_{AF} should increase exponentially with a large exponent and, therefore, the CPP GMR ratio defined by Eq. (5) should also grow exponentially with N.

These general arguments were already presented in Ref. 1 and illustrated by model calculations for a single-orbital tight-binding band. However, the pertinent question is whether the localization lengths in the AF channel are short enough in real systems so that localization can influence the CPP GMR. This question can only be answered by an exact evaluation of the Kubo formula for specific Co/Cu and Fe/Cr pseudorandom spin valves. The formalism developed in Secs. II and III remains valid in the Anderson localization regime and can be readily applied to Co/Cu and Fe/Cr pseudorandom spin valves. I have made such calculations for $Co_{2-6}Cu_{5-8}$ and $Fe_{2-10}Cr_{4-10}$ valves and their CPP GMR ratios are plotted on a logarithmic scale against the number of bilayers N in Fig. 7 (squares for Co/Cu and circles for Fe/Cr). The thicknesses of Cu layers in the Co/Cu valve were made to fluctuate between 5 and 8 atomic planes. These limits were imposed so that the interlayer exchange coupling remains antiferromagnetic. For the same reason, the thicknesses of Cr layers were made to fluctuate between 4 and 10 atomic planes. There is no real restriction on the range of fluctuations in thickness of the ferromagnetic layers. However, for computational reasons (convergence of the BZ sum), the total thickness of the valve cannot exceed $\approx$150 nm. I have, therefore, restricted rather arbitrarily the mean thickness of Co layers to 4 atomic planes ($M_{\min}=2$, $M_{\max}=6$) and the mean thickness of Fe layers to 6 atomic planes ($M_{\min}=2$, $M_{\max}=10$).

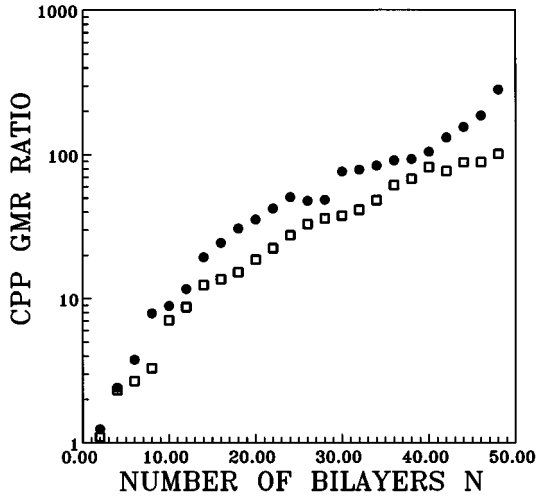

FIG. 7. CPP GMR ratios of $Co_{2-6}Cu_{5-8}$ (squares) and $Fe_{2-10}Cr_{4-10}$ (circles) pseudorandom spin valves plotted on a logarithmic scale against the number of bilayers N.

As expected, the CPP GMR increases approximately exponentially with the number of bilayers N both for the Co/Cu and Fe/Cr pseudorandom valves. The maximum R_{CPP} achieved for the Fe/Cr valve with $N=50$ is approximately 3×10^4 %, which is about two hundred times greater than the maximum R_{CPP} observed in the Ohmic regime.[4] This is, of course, not the upper theoretical limit but merely a limit imposed by the computer time available. The theoretical CPP GMR increases with increasing number of bilayers without any saturation as long as the valve remains in the localization regime.

To understand the precise reason for such a large enhancement of the CPP GMR, one needs to examine the individual resistances in the FM and AF configurations. The resistances $R_{\mathrm{FM}}^{\uparrow}$, $R_{\mathrm{FM}}^{\downarrow}$, and $R_{\mathrm{AF}}^{\uparrow,\downarrow}$ for the Co/Cu valve are plotted in Fig. 8 on a logarithmic scale against N. The corresponding results for the Fe/Cr valve are shown in Fig. 9. Consider first the Co/Cu valve. The resistance in the AF configuration (circles in Fig. 8) and the resistance in the down-spin channel in the FM configuration (triangles) increase approximately exponentially with N due to Anderson localization. Their values for $N=50$ are, therefore, a factor of twenty larger than for a superlattice in the Ohmic regime (Figs. 5 and 6). On the other hand, the resistance in the up-spin channel in the FM configuration (squares) remains as low as in the Ohmic regime. The reason for this behavior is that localization does not set in for such small N in this channel because disorder is almost negligibly weak due to excellent matching of the up-spin bands in Co to the Cu

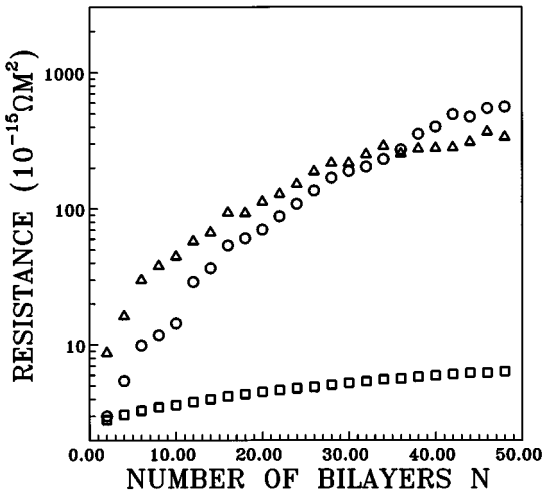

FIG. 8. Resistances in the ferromagnetic (FM) and antiferromagnetic (AF) configurations of a $Co_{2-6}Cu_{5-8}$ pseudorandom spin valve plotted on a logarithmic scale against the number of bilayers N. Circles denote the resistance $R_{AF}^{\uparrow,\downarrow}$; squares the resistance $R_{FM}^{\uparrow}$ and triangles the resistance $R_{FM}^{\downarrow}$.

bands. It follows that the very large enhancement of the CPP GMR for the Co/Cu pseudorandom spin valve is due, entirely, to the Anderson localization of electrons in the AF configuration.

The magnetic contrast of the Fe/Cr pseudorandom valve is even more enhanced by the Anderson localization. The resistances for $N=50$ in the AF configuration and in the up-spin channel in the FM configuration are two orders of magnitude greater than the corresponding resistances in the Ohmic regime (Fig. 6), whereas the resistance in the down-spin channel in the FM configuration is virtually unaffected by disorder. The reason is, of course, a very good matching of Fe and Cr bands in the down-spin channel.

An exact numerical evaluation of the Kubo formula for Co/Cu(001) and Fe/Cr(001) pseudorandom spin valves using

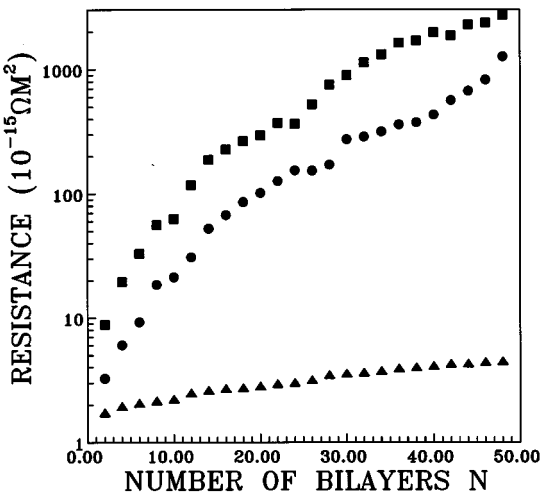

FIG. 9. Resistances in the ferromagnetic (FM) and antiferromagnetic (AF) configurations of a $Fe_{2-10}Cr_{2-10}$ pseudorandom spin valve plotted on a logarithmic scale against the number of bilayers N. Circles denote the resistance $R_{AF}^{\uparrow,\downarrow}$; squares the resistance $R_{FM}^{\uparrow}$; and triangles the resistance $R_{FM}^{\downarrow}$.

an *ab initio* band structure of the constituent metals thus confirms the very large enhancement of the CPP GMR predicted in Ref. 1. The enhancement is due to multiple scattering of electrons in the AF configuration from a highly disordered sequence of ferromagnet/spacer interfaces. Such scattering gives rise to electron localization with short localization lengths of the order of twenty bilayers (30–40 nm).

As in Sec. III, a comment on the nature of randomness in pseudorandom spin valves is required. The situation for a pseudorandom spin valve is qualitatively different from that for a superlattice with small spontaneous fluctuations in layer thickness. Once a pseudorandom spin valve is prepared with layer thicknesses following a predetermined pseudorandom number sequence, 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 layers with the same known pseudorandom sequence, and vice versa. In other words, the relevant quantity to be calculated is the sample specific GMR and, therefore, any configuration averaging over disorder of the interfaces would be completely inappropriate.

V. CONCLUSIONS

The conventional explanation of the GMR effect is based on spin-dependent scattering of electrons from magnetic impurities located at ferromagnet/spacer interfaces (interfacial spin-dependent scattering). The resulting transport problem is solved either within the classical Boltzmann formalism[21] or within a linear response theory with a simplified band structure (parabolic bands).[22] Realistic modelling of interfacial roughness combined with a rigorous quantum evaluation of the CPP GMR from the Kubo formula was made by Asano *et al.*[11] but only for a single-orbital tight-binding band structure. More recently, Butler *et al.*,[23] Zahn *et al.*[24], and Nesbet[25] solved the Boltzmann equation with a fully realistic band structure. However, a common feature of all these theories, with the exception of Ref. 11, is that the GMR effect disappears when the spin-dependent impurity scattering is switched off. Since interfacial impurity scattering is linked directly to interfacial roughness, the implication of all the above theories is that the GMR effect vanishes (or is negligibly small[26]) for perfectly flat interfaces.

This conventional point of view was challenged by Schep *et al.*[3] They considered the simplest case of GMR without impurity scattering, i.e., an infinite perfectly periodic superlattice. Using an *ab initio* band structure, they obtained CPP GMR ratios in excess of 100 %. These very high values of CPP GMR are due entirely to quantum scattering from perfectly flat interfaces.

In this paper, I have included an additional important ingredient, i.e., fluctuations in layer thickness, and investigated comprehensively the CPP GMR due to scattering from otherwise perfectly flat interfaces (GMR without impurity scattering) for Co/Cu(001) and Fe/Cr(001) finite superlattices sandwiched between two semi-infinite contacts. Using an *ab initio* band structure for all the constituent metals and solving the quantum transport problem exactly (numerical evaluation of the Kubo formula), I find that CPP GMR without impurity scattering is far from negligible and can easily explain the whole observed effect. Moreover, depending on the size of

fluctuations in layer thickness, CPP GMR without impurity scattering occurs in three distinct regimes: ballistic, Ohmic, and Anderson localization (pseudorandom spin valve).

When there are no fluctuations in layer thickness, CPP transport is in the ballistic regime. The ballistic CPP GMR ratio R_{CPP} of a finite superlattice saturates rapidly as a function of the number of bilayers (only ≈ 5 bilayers are needed) and reaches a value equal to R_{CPP} for an infinite superlattice. The saturation values of R_{CPP} obtained from the Kubo formula are of the order of 100 % both for Co/Cu and Fe/Cr superlattices, which is in a very good agreement with the results obtained earlier by Schep *et al.*[3,6] for infinite Co/Cu and Fe/Cr superlattices. However, the fact that the absolute value of the resistance of a superlattice in the ballistic regime is far too low compared with the experiment[4] and also that R_{CPP} saturates so rapidly clearly indicates that ballistic effects are not seen in present experiments.

When small fluctuations in layer thickness corresponding to only one atomic plane at the interface being displaced are introduced, transport changes from ballistic to Ohmic. The calculated GMR ratio R_{CPP} increases initially linearly with the number of bilayers N and then saturates for $N \approx 40-50$. Such a behavior is a signature of the Ohmic regime and was observed for Co/Ag and Co/Cu by Schroeder *et al.*[4] The maximum calculated saturation values of R_{CPP} are in the region 800–1000 %. However, much more significantly, the absolute values of the zero-field (AF) and saturation-field (FM) resistances calculated without any adjustable parameters increase linearly with N (good Ohm's law), which is as observed,[4] and the zero-field (AF) resistance of the Co/Cu superlattice of the same thickness and composition as the Co/Cu(111) sample investigated in Ref. 4 has a value of $165 \times 10^{-15}\ \Omega\ \mathrm{m}^2$, which is within 10 % of the observed resistance.

These results indicate very strongly that it is scattering from fluctuations in layer thickness rather than the conventional interfacial scattering that determines the CPP GMR observed in present experiments. Further support for this mechanism comes from an analysis of x-ray scattering data[27] which shows that fluctuations in layer thickness of the order of one atomic plane are always present in experimental samples. One can go even further and argue that, without fluctuations in layer thickness, the observed CPP GMR cannot be explained at all. This is because fluctuations in layer thickness *increase* the CPP GMR ratio whereas interfacial roughness *decreases* it. The latter result was proved quite rigorously by Asano *et al.*[11] for a single-orbital tight-binding band. They showed that CPP GMR ratio has its maximum value for perfectly flat interfaces and is always *reduced* from the maximum value when ferromagnet and spacer atoms are intermixed at the interface. Given that interfacial roughness is detrimental for CPP GMR and *ab initio* calculations for periodic Co/Cu superlattices with perfectly flat interfaces give an upper limit on CPP GMR of about 120–150 %, which is smaller than the maximum observed[4] effect of about 170 %, it is clear that, without fluctuations in layer thickness, the magnitude of the observed CPP GMR cannot be explained. One may, therefore, conclude that fluctuations in layer thickness is an important source of CPP GMR that has not been considered in previous theoretical treatments and the fact that it is required to account quantitatively for the observed CPP GMR effect must be significant.

The only discrepancy between the calculated and observed results in the Ohmic regime is that the calculated saturation-field (FM) resistance is a factor of three smaller than the observed value. One possible explanation is that there is some additional scattering mechanism, other than spin-dependent scattering from interfaces, which is not included in the present calculation. However, a more likely explanation is that the matching between the Co majority and Cu bands (Fe minority and Cr bands), which determines the saturation-field resistance, may not be so perfect when lattice relaxation effects are included. In fact, the saturation-field resistance calculated with bulk Co and Cu parameters is almost certainly underestimated. The matching in the up-spin channel for bulk Co and Cu bands is so perfect that any modification of the band structure due to lattice relaxation is bound to make the matching poorer, and hence, increase the saturation-field resistance. This would, at the same time, bring down somewhat the calculated CPP GMR and, hence, make the agreement between the theory and experiment better.

When large fluctuations in layer thickness are introduced deliberately, the Ohmic regime changes into, experimentally as yet unexplored, Anderson localization regime in which the proposed pseudorandom spin valves[1] operate. If high-quality Co/Cu and Fe/Cr pseudorandom spin valves could be fabricated, the results of Sec. IV show that they would have CPP GMR ratios at least a factor hundred higher than the highest currently attainable values.

Successful operation of Co/Cu and Fe/Cr pseudorandom spin valves depends on two conditions: (i) the saturation-field (FM) resistance of a pseudorandom spin valve must not be much higher than the calculated resistance; (ii) the zero-field (AF) resistance must be sufficiently enhanced by Anderson localization.

The first condition is easy to satisfy. The FM resistance R_{FM} of present Co/Cu superlattices is only a factor 3 higher than the calculated R_{FM}. The results of Secs. III and IV show that disordering the sequence of interfaces has virtually no effect on R_{FM}. There is, therefore, no reason to expect that R_{FM} of a carefully prepared Co/Cu pseudorandom spin valve should be any higher than R_{FM} in the present Ohmic regime.

For a pseudorandom spin valve with all dimensions smaller than the mean free path, the calculations of Sec. IV are exact for real Co/Cu and Fe/Cr systems and the second condition is, therefore, also satisfied. It follows from Fig. 7 that CPP GMR of about 5000 % should be achieved for a valve of about 50 nm thick. The thickness of the valve is, therefore, not a serious problem since 50 nm is comparable to the mean free path. One should clearly try to keep all the layers as thin as possible while maintaining large fluctuations in their thickness in order to squeeze as many bilayers as possible into a total thickness comparable to the mean free path. The only question that remains unresolved is whether the transverse dimensions of the valve must also be small, i.e., comparable to the mean free path. One can argue that impurity scattering in a macroscopic valve would lead to mixing of $\vec{k}_{\parallel}$ channels which might eventually destroy one-dimensional localization. However, what the upper bound is

on the valve diameter, if any, is difficult to estimate theoretically.

Finally, all the calculations presented here are at zero temperature. Inelastic scattering at finite temperatures might also spoil the localization in the AF configuration and thus reduce the calculated GMR ratios. However, the recent results of Pascual *et al.*[28] for very thin gold wires are encouraging in this respect since they observed Anderson localization in disordered one-dimensional gold wires even at room temperature. One can, therefore, conclude that the predicted very large enhancement of the CPP GMR in pseudorandom spin valves should certainly be observable in Co/Cu and Fe/Cr valves with all dimensions smaller than the mean free path at low temperatures. It is, however, quite likely that a substantial enhancement persists in valves with more macroscopic transverse dimensions and also at finite temperatures.

ACKNOWLEDGMENTS

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