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Giant tunnel magnetoresistance in multilayered metal/oxide structures comprising multiple quantum wells

A Vedyayev†‡, N Ryzhanova‡, R Vlutters†§ and B Dieny†

† CEA, Département de Recherche Fondamentale sur la Matière Condensée, SP2M/NM, 38054 Grenoble Cédex 9, France

‡ Department of Physics, Moscow Lomonosov University, Moscow 119699, Russia

§ MESA Research Institute, University of Twente, PO Box 217, 7500 AE Enschede, Netherlands

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Abstract. From a theoretical point of view, we have investigated the transport properties in metal/oxide multilayer structures of the form M/O/M/O/M/O/M where M represent ferromagnetic layers alternating with three insulating barriers (O = oxide). The two inner magnetic layers form two quantum wells, the depths of which are spin dependent. For particular thicknesses of these magnetic layers, resonances occur in the quantum wells which lead to strong increase in the electron transmission through the insulating barriers. We show that if the magnetization in the successive magnetic layers can be changed from parallel to antiparallel as in spin-valves, then, for particular values of the thicknesses of the two inner magnetic layers, very large magnetoresistance effects can be expected due to the interplay of resonance effects in the two neighbouring quantum wells. The conductivity and magnetoconductivity are calculated within a quantum theory of linear response (Kubo formalism) taking into account the scattering in the magnetic layers. We show that in such a structure, giant tunnel magnetoconductivity can arise not only from a difference between spin up and spin down Fermi wave-vectors in the magnetic layers but also from spin-dependent mean free paths. In the latter case, the effect of the scattering is to induce a spin-dependent broadening of the resonances in the quantum wells.

The recent observations of large magnetoresistance (MR) effects at room temperature [1–3] in tunnel junctions of the form MOM′ (M and M′ = magnetic metals, O = oxide tunnel barrier) [4] has stimulated a renewed interest for these systems, named magnetic valves [5]. Besides their fundamental interest, these structures are foreseen as being potential candidates for very sensitive magnetic field sensors or as memory cell in magnetic random access memories. The largest MR amplitudes have been observed so far, in Al/Al_2O_3 junctions prepared by oxygen plasma oxidation [1–3].

In magnetic valves, the tunnel conductance varies as a function of the angle θ between the magnetizations in the two ferromagnetic layers (magnetic valve effect). So far, the experimental results on these systems have been interpreted on the basis of Slonczewski's theory [5]. This author calculated the conductance of a MOM′ junction in a free electron model taking into account the exchange splitting of the d band. The calculation was made in the framework of classical quantum mechanics. No scattering of electrons in the magnetic metallic electrodes was taken into account.

Recently, we showed that spin-dependent antireflective coating effects can occur in tunnel junctions of the form MPOPM′ in which thin paramagnetic layers (P) are inserted at the interface between the magnetic and the non-magnetic layers [6]. For certain choices of the

(M, P) couple, for example (Co, Cu), it has been shown, from the studies on oscillatory coupling in (Co/Cu) multilayers, that a good matching of the majority-electron-spin band occurs at the Fermi energy between Co and Cu whereas the minority electrons feel a significant potential step at the Co/Cu interface. The resulting spin-dependent reflection coefficient at Co/Cu interfaces is believed to be at the origin of the oscillatory coupling in these multilayers [7]. In a similar way, in a $Co/Cu/Al_2O_3/Cu/CoFe$ junction, the Cu layers act, for spin down electrons, as quantum wells adjacent to the barrier. When the Fermi energy coincides with a quantum well level, a resonance effect occurs in which the incident electrons undergo multiple reflections on the edges of the well. These reflections lead to a very strong enhancement of the transmission of spin down electrons through the barrier. Since this effect does not occur for spin up electrons, it results in a strong enhancement in the effective polarization of the tunnel electrons and therefore in a strong increase in the MR amplitude.

Zhang *et al* theoretically investigated the transport properties through double tunnel junctions of the form MOMOM [8]. In such structure, the central magnetic layer M constitutes a spin-dependent quantum well in which resonance effects occur when the width of the well is an integer number of the Fermi wavelength. The authors showed that at resonances the MR ratio in these systems can be 3 to 4 times larger than in usual MOM′ magnetic valves. Furthermore, the conductivity through the system enhances dramatically at the resonances [8]. However, in such a structure containing only one quantum well, the occurrence of the resonant tunnelling throughout the structure does not depend on the relative orientation of the magnetization in the successive magnetic layers. Consequently, the conductivity of such structure increases dramatically at resonances but not the magnetoresistance. Furthermore, in [8], the width of the resonance was assumed to be very small (proportional to $e^{-2q_0 b}$ where q_0 is the damping vector of the evanescent wave within the barrier and b is the barrier width) because the scattering in the metallic layers was neglected. However, in real systems, electron mean free paths in magnetic metallic layers are of the order of one to a few nanometres which is not much longer than the typical layer thickness. This means that electron scattering can play a significant role and must be included in the calculation of the transport response of these systems. The main effect of the scattering is to decrease the sharpness of the resonances which has strong consequences on the resonant tunnelling.

In this paper, we consider the transport properties across a more complex multilayered structure of the form $M_1OM_2OM_3OM_4$ consisting of two thin magnetic layers (M_2 and M_3) inserted between three tunnel barriers with two magnetic electrodes at the edges of the structure (M_1 and M_4). We assume that, somehow, it is possible to change the relative orientation of the magnetization in the successive ferromagnetic layers from parallel to antiparallel as in giant-magnetoresistance multilayers. This can be achieved, for instance, by using ferromagnetic layers of different coercivities. In such a structure, we show that for particular thicknesses of the magnetic layers, a very large change in tunnel conductivity can occur between the parallel and antiparallel magnetic configurations. Indeed, M_2 and M_3 constitute two quantum wells, the depths of which are spin dependent. In the parallel magnetic configuration, it is possible to choose the thickness of M_2 and M_3 in such a way that resonances occur in both quantum wells for spin up electrons. For that purpose, the thicknesses of the two layers must be integer numbers of the spin up Fermi wavelengths. The occurrence of simultaneous resonances in the two adjacent wells leads to a very strong increase in the overall tunnel conductivity. When the magnetic configuration switches to antiparallel, the resonance occurs in only one of the layers. As a result, the overall tunnel conductivity becomes much lower. This contrasts with the situation of single quantum well MOMOM structures [8] in which the resonant tunnelling is not affected by the relative orientation of the magnetization in the successive magnetic layers. More quantitatively, we can define a dimensionless width of resonance Γ_σ (σ refers to the spin of the electrons) which is influenced by the scattering in the quantum well and the probability of transmission through the tunnel barriers. In most practical cases, the former contribution is dominant so that $\Gamma_\sigma \approx (a/l_\sigma) \ll 1$ where a is the width of the quantum well and l_σ the elastic mean free path. It can be shown that the ratio of conductivities between parallel and antiparallel configurations is then proportional to $(\Gamma_\sigma)^{-2} \gg 1$. Therefore, this triple barrier–double magnetic quantum well structure is a real resonant tunnel magnetic valve device. Very large magnetoresistance effects can be expected in such structures, much larger than in the double junction systems studied in [8].

We now describe in more detail our model. We consider the system M/O b/M a/O b/M a/O b/M in which we assume for simplicity that all magnetic layers are made of the same magnetic material. The two inner magnetic layers have the same thickness a. The oxide barriers O have a thickness b. The outer electrodes are supposed to be semi-infinite. As in [5], the electrons are assumed to form a free electron gas with exchange splitting of spin subbands in the ferromagnetic layers. We take into account the scattering in the metallic layers by introducing the elastic mean free paths, which are spin dependent in the magnetic layers (l_1 and l_2 respectively represent the spin up and spin down mean free paths in M). We then calculate the conductivity of this structure in the linear regime by using Kubo formalism (Green function method). This approach has been described in our previous paper [6]. The final expressions of the conductances in parallel ($\sigma^{\uparrow\uparrow}$) and antiparallel ($\sigma^{\uparrow\downarrow}$) magnetic configurations are given by

$$\begin{aligned}\sigma^{\uparrow\uparrow} = \frac{e^2 q_0^2}{4\hbar\pi}\Bigg\{&\Bigg[\left(\int_0^{\left(\frac{k_1^F}{q_0}\right)^2} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_1)\Phi(\kappa_1)\,\mathrm{d}x\right)^{-1} \\ &+\frac{1}{2}\left(\int_0^{\left(\frac{k_1^F}{q_0}\right)^2} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_1)\Psi(\kappa_1)\,\mathrm{d}x\right)^{-1}\Bigg]^{-1} \\ &+\Bigg[\left(\int_0^{\left(\frac{k_2^F}{q_0}\right)^2} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_2)\Phi(\kappa_2)\,\mathrm{d}x\right)^{-1} \\ &+\frac{1}{2}\left(\int_0^{\left(\frac{k_2^F}{q_0}\right)^2} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_2)\Psi(\kappa_2)\,\mathrm{d}x\right)^{-1}\Bigg]^{-1}\Bigg\} \end{aligned} \tag{1}$$

and

$$\begin{aligned}\sigma^{\uparrow\downarrow} = \frac{e^2 q_0^2}{2\hbar\pi}\Bigg[&\left(\int_0^{Min} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_1)\Phi(\kappa_2)\,\mathrm{d}x\right)^{-1} + \left(\int_0^{Min} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_1)\Psi(\kappa_2)\,\mathrm{d}x\right)^{-1} \\ &+\left(\int_0^{Min} q^2\,\mathrm{e}^{-2qb}\Psi(\kappa_2)\Phi(\kappa_1)\,\mathrm{d}x\right)^{-1}\Bigg]^{-1}\end{aligned} \tag{2}$$

with

$$\mathrm{Min} = \mathrm{Min}\left(\left(\frac{k_1^F}{q_0}\right)^2, \left(\frac{k_2^F}{q_0}\right)^2\right) \qquad \Psi(\kappa_i) = \frac{c_i \sinh 2d_i a(q^2 + c_i^2)}{\mathrm{Den}_i} \tag{3}$$

$$\begin{aligned}\Phi(\kappa_i) = [\![c_i\{(c_i^2+q^2)^2\cosh 2d_i a - [(q^2-c_i^2)\cos c_i a - 2c_i q\sin c_i a]^2 \\ +[(q^2-c_i^2)\sin c_i a + 2c_i q\cos c_i a]^2\}]\!][\![(c_i^2+q^2)\mathrm{Den}_i]\!]^{-1}\end{aligned} \tag{4}$$

$$\begin{aligned}\mathrm{Den}_i = \sinh^2 d_i a[(q^2-c_i^2)\cos c_i a - 2c_i q\sin c_i a]^2 + \cosh^2 d_i a[(q^2-c_i^2)\sin c_i a \\ +2c_i q\cos c_i a]^2\end{aligned} \tag{5}$$

where $q = q_0\sqrt{1+x}$, and $\mathrm{i}\hbar q_0 = \mathrm{i}\sqrt{2m(u_0-\varepsilon_F)}$ represents the imaginary electron momentum in the barrier. $(u_0-\varepsilon_F)$ is the barrier height relative to the Fermi energy. The variable of integration in (1) and (2) is $x = \kappa^2/q_0^2$ where κ is the component of the electron momentum parallel to the plane of the layers. k_i can be written as $k_i = c_i + d_i = \sqrt{(k_i^F)^2 - q_0^2 x + (2k_i^F/l_i)\mathrm{i}}$. k_1^F and k_2^F are the Fermi momenta of respectively spin up and spin down electrons in the ferromagnetic layers. In expression (3), it is easy to show that the factor $\Psi(\kappa_i)$ oscillates as a function of the thickness a of the ferromagnetic layers and exhibits resonances for particular thicknesses a_0^i given by

$$\tan(a_0^i c_i) = \frac{2qc_i}{c_i^2 - q^2}. \tag{6}$$

In the limit $a/l_i \ll 1$, the amplitude of this factor at resonances can be written as:

$$\Psi(\kappa_i)|_{a=a_0^i} \approx \frac{2c_i a_0^i/l_i}{(q^2+c_i^2)[(a_0^i/l_i)^2 + c_i^2(a-a_0^i)^2]}. \tag{7}$$

The width of the resonances is $c_i^{-1}a_0^i/l_i$.

We then estimate the magnetoresistance amplitude of this multiple junction system when the thickness of the magnetic layers corresponds to a resonance, from the asymptotic values of integrals (1) and (2), assuming $q_0 b \gg 1$. We find

$$\mathrm{MR}_{max} = \frac{(l_i/a)(l_j/a)(q^2+k_j^F)^2(k_j^F)^2(a_0^i - a_0^j)^2}{4(q^2+k_i^F)^2} \qquad (i \neq j). \tag{8}$$

Because of the large possible values of the product $l_i l_j/a^2$, great enhancement of MR amplitudes around resonances can be achieved in these structures.

Another interesting point is that even if the Fermi momenta of spin up and spin down are equal, large magnetoresistance effects can still be obtained due to the spin-dependent scattering of electrons in the magnetic layers. For $k_1^F = k_2^F$ and $l_1 \neq l_2$, the MR amplitude can reach the maximum value

$$\mathrm{MR}_{max} \approx (l_1 - l_2)^2/4l_1 l_2 \tag{9}$$

which has the same expression as the giant magnetoresistance amplitude expected for metallic spin-valve multilayers in the current-perpendicular-to-the-plane (CPP) geometry without spin-flip scattering [9].

To illustrate the obtained results, we have calculated the dependencies of the conductivities and MR ratio $(\sigma^{\uparrow\uparrow} - \sigma^{\uparrow\downarrow})/\sigma^{\uparrow\downarrow}$ versus the thickness of the two ferromagnetic layers in three cases: (i) the MR originates from different Fermi wave vector for spin up and spin down electrons in the magnetic material, (ii) the Fermi wave vectors are equal but the mean free paths are spin dependent, (iii) both Fermi wave vectors and mean free paths are spin dependent. In these calculations, the considered structure is F/O 10 Å/F a Å/O 10 Å/F a Å/O 10 Å/F in which the two outer layers are thick so that no quantum size effects arise from these layers. For the first case, we chose $l_1 = l_2 = 100$ Å and $k_1^F = 1$ Å^{-1}, $k_2^F = 0.4$ Å^{-1} (appropriate values for itinerant d electrons in Fe [10]), $q_0 = 1$ Å^{-1} (corresponding to typical barrier height for Al_2O_3). Figure 1 and its inset respectively show the magnetoresistance and spin-dependent conductivities of the structure. Well defined oscillations occur for spin up and spin down conductivities as a function of the width of the quantum well states. The period of these oscillations is half the Fermi wavelength of the corresponding electrons. In the parallel magnetic configuration, resonances occur in the conductivity whenever the Fermi energy simultaneously matches quantum well levels in the two adjacent wells. In antiparallel magnetic configuration, this simultaneous matching does

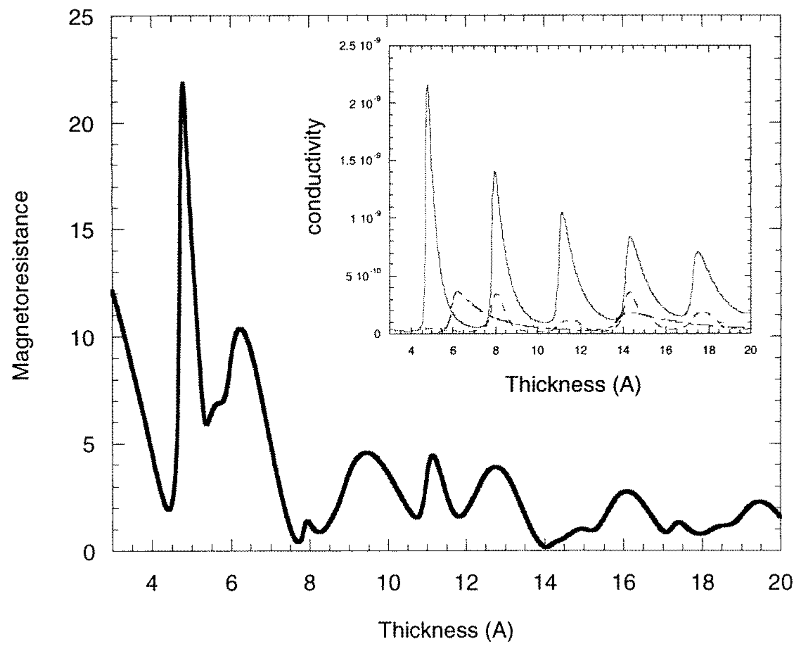

Figure 1. Magnetoresistance ratio of a triple barrier–double quantum well system of the composition F/O 10 Å/F a Å/O 10 Å/F a Å/O 10 Å/F. $l_1 = l_2 = 100$ Å; $k_1^F = 1$ Å^{-1}, $k_2^F = 0.4$ AA^{-1}; $q_0 = 1$ Å^{-1}. Inset: spin-dependent conductivities: —— spin up electrons in parallel magnetic configuration; – – – spin down electrons in parallel magnetic configuration; - - - - spin up and down electrons for antiparallel magnetic configuration.

not occur since the thicknesses of the two inner magnetic layers are assumed to be equal and $k_1^F \neq k_2^F$, resulting in a much lower conductivity. The damping of the oscillations is due to the scattering in the magnetic layer. The characteristic length associated with this damping is the elastic mean free path (l_σ) [6]. The magnetoresistance exhibits some complicated oscillations. At the first resonance, the MR reaches 2000%. This maximum value is lower than derived from the approximate expression (8). This discrepancy originates from the fact that expression (8) is obtained in the limit $x = \kappa^2/q_0^2 \to 0$ by considering that the function to integrate decreases very sharply with x because of the e^{-2qb} factor. However since the function $\Psi(\kappa_i)$ exhibits resonances in the integration region for $x \neq 0$, this approximation is not very appropriate and a numerical integration as used in figure 1 is more reliable. In a real system, the oscillations would probably be smeared out due to spatial fluctuations in the layer thickness caused by interfacial roughness. However, the average amplitude remains fairly large, of the order of 500% for thicknesses below 8 Å or 300% between 8 and 14 Å. Therefore, it should be possible to observe these large MR effects experimentally.

In the second case (figure 2), we assumed $k_1^F = k_2^F = 1$ Å^{-1} and $l_1 = 100$ Å, $l_2 = 10$ Å. This would represent the case where the conduction by slightly exchange split s-electrons is dominant. The oscillations of conductivities are in phase for the two species of electrons. The effect of the spin-dependent scattering is to lead to different broadening of the resonances in conductivity for spin up and spin down electrons (see inset of figure 2). The MR amplitudes are lower than in the former case. However, the peaks in MR are broader than in the former case which may make them more easily observable from an experimental point of view. The maximum MR amplitude coincides here nicely

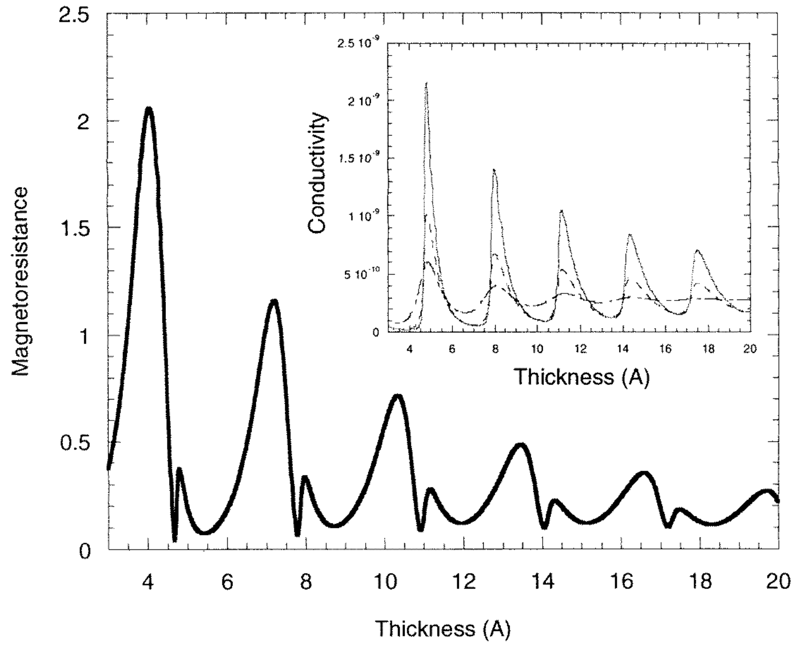

Figure 2. Magnetoresistance ratio of a triple barrier–double quantum well system of the composition F/O 10 Å/F a Å/O 10 Å/F a Å/O 10 Å/F. $l_1 = 100$ Å, $l_2 = 10$ Å; $k_1^F = k_2^F = 1$ Å^{-1}; $q_0 = 1$ Å^{-1}. Inset: spin-dependent conductivities: —— spin up electrons in parallel magnetic configuration; – – – spin down electrons in parallel magnetic configuration; - - - - spin up and down electrons for antiparallel magnetic configuration.

with the result of expression (9) ($MR_{max} \approx 2$ from expression (9)). This amplitude is the same than for CPP MR in metallic multilayers but with a much larger CPP resistance.

In figure 3, the two previous situations are combined: $k_1^F = 1$ Å^{-1}, $k_2^F = 0.4$ Å^{-1} and $l_1 = 100$ Å, $l_2 = 10$ Å. This case is closer to experimental situations. The results are intermediate between the two previous cases. The MR amplitudes are large and the maxima in MR may be large enough to be observed experimentally.

We now briefly discuss the effect that interfacial roughness, inherent to real systems, may have on the observation of the previously discussed transport properties. Three cases should be distinguished: (i) In case of a correlated roughness, each interface exhibit atomic steps but the location of these steps is the same for all interfaces. The thicknesses of the two inner magnetic layers is then uniform so that the magnetoresistance should not be much affected compared to the ideal case. (ii) If the roughness is uncorrelated, then the thicknesses of M_2 and M_3 fluctuate independently of each other. The resonances may no longer occur simultaneously in the two quantum wells which would significantly reduce the magnetoresistance amplitude. (iii) The roughness may also be at the atomic scale because of interdiffusion between adjacent layers. In that case, the diffuse scattering of electrons at the interface can slightly change the position of the resonance and give an additional contribution to its width. The bulk scattering leads to a resonance width proportional to a/l whereas, as we will show in [11], the interfacial scattering gives a contribution proportional to $(k^F\lambda)^{-1}$ where λ is the characteristic length associated with interfacial scattering. For sufficiently smooth interface, this additional contribution is lower than the bulk one so that

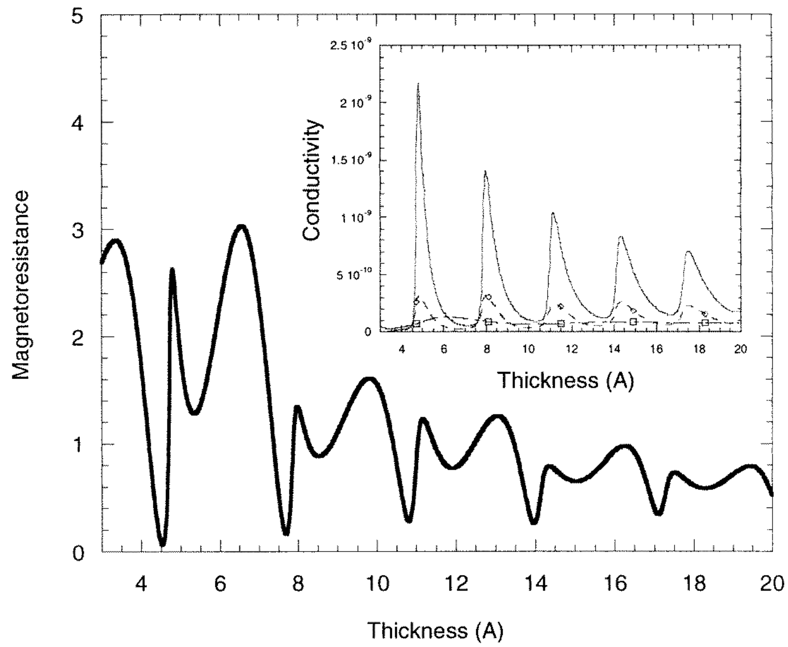

Figure 3. Magnetoresistance ratio of a triple barrier–double quantum well system of the composition F/O 10 Å/F a Å/O 10 Å/F a Å/O 10 Å/F. $l_1 = 100$ Å; $l_2 = 10$ Å; $k_1^F = 1$ Å^{-1}; $k_2^F = 0.4$ Å^{-1}; $q_0 = 1$ Å^{-1}. Inset: spin-dependent conductivities: —— spin up electrons in parallel magnetic configuration; – – – spin down electrons in parallel magnetic configuration; - - - - spin up and down electrons for antiparallel magnetic configuration.

the observed properties are not much affected by the interfacial scattering. In the opposite case, a significant broadening of the resonances may occur, thus leading to a reduction in conductivities and MR amplitude.

In conclusion, we have shown that very large magnetoresistance effects can be expected in triple barrier–double quantum well magnetic systems (MOMOMOM). These effects arise from the possibility to tune the position of the quantum levels in the two adjacent quantum wells by changing the relative orientation of the magnetization in the successive magnetic layers. The idea developed in this paper could be extended to more complex multilayered structures of the form M/(O/M)$_n$ ($n > 3$) with even larger MR amplitude.

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