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Interpretation of the magnetoresistance in doped magnetic tunnel junctions

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Abstract. We present a quantum mechanical model of the magnetoresistance in ferromagnetic tunnel junctions artificially doped by the introduction of layers of impurities in the middle of the barrier. The electron transport across the barrier is described by a combination of direct tunneling, tunneling assisted by spin-conserving scattering and tunneling assisted by spin-flip scattering. With this model, we interpret recent experimental results concerning the dependence of the TMR amplitude on the amount of impurities in the barrier and on temperature.

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Magnetic tunnel junctions consist of two ferromagnetic layers separated by an insulating barrier. By using two ferromagnetic materials of different coercivity or by pinning the magnetization of one of the ferromagnetic layer by exchange anisotropy, the relative orientation of the magnetization in the two magnetic electrodes can be changed from parallel to antiparallel by an external applied field. The TMR amplitude is defined as the ratio $\frac{\sigma^{\mathrm{P}}-\sigma^{\mathrm{AP}}}{\sigma^{\mathrm{AP}}}$, where $\sigma^{\mathrm{P(AP)}}$ is the conductance of the structure for parallel (antiparallel) magnetic configuration. Recently, Jansen and Moodera investigated the influence on the magnetoresistance (TMR) of Co/Al_2O_3/NiFe magnetic tunnel junctions of the presence of impurities (Co, Ni, Pd, Cu) artificially introduced in the middle of the tunnel barrier [1]. It was observed that the TMR decreases approximately linearly with increasing impurity content. The decrease is most pronounced for Ni, relatively weak for Co and intermediate for Cu and Pd. A correlation was found between the rate of the decrease and the spin carried by the impurities embedded in Al_2O_3. These results showed that the TMR can be severely reduced as a result of spin-flip scattering in the barrier.

From a theoretical point of view, Tsymbal *et al.* investigated the influence of impurities within disordered insulators on the spin polarization of tunneling electrons and on the magnetoresistance [2]. The disorder in the insulator was introduced through a randomness of the onsite energies of the insulator host atoms. Furthermore, the impurities were assumed to be randomly distributed inside the barrier with a fixed onsite energy. Spin-flip processes were not taken into account. The conductance of this type of tunnel junctions is mainly determined by two mechanisms: (i) Tunneling through resonant channels, specific for each disordered barrier, (ii) Tunneling *via* impurity mini-bands, which are formed within the band gap of the insulator. In reference [2], it was found that the latter mechanism increases the conductance by several orders of magnitude when 5% impurities are introduced. However in the experiments of reference [1], the introduction of impurities results in a decrease of the conductance only by a factor of two. This relatively weak conductance variation is due to the fact that all the impurities introduced within the Al metallic layer are also oxidized during the oxidation of Al in Al_2O_3. Therefore the impurities in [1] must be considered as additional barriers for the tunneling electrons but with a barrier height which can be different from that of alumina. In this case, in contrast to the situation investigated in reference [2], there are no resonant states inside the barrier. The difference in the heights of the host and impurity barriers leads to scattering of the tunneling electrons. The resonant tunneling which was considered in reference [2] does not occur here.

In this paper, we present a quantum theory of TMR in doped junctions such as those studied in reference [1]. The theory takes into account three tunneling processes across the barrier: direct tunneling, tunneling assisted by spin-conserving scattering and tunneling assisted by spin-flip scattering. This model allows us to interpret Jansen and Moodera's results [1] with two adjustable parameters that will be described further.

Our model system is a sandwich of the form F1/O/F3. F1 and F3 are two semi-infinite ferromagnetic layers and O

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is the insulating tunnel barrier of thickness b. Furthermore, we assume that a submonolayer of impurities is introduced in the middle of the barrier. These impurities are first characterized by their concentration $c_{\rm B}$ in this submonolayer plane. The atomic concentration $c_{\rm B}$ is given by $c_{\rm B} = \frac{t_{\rm impurity}}{a_{\rm Al_2O_3}}$ where $t_{\rm impurity}$ is the effective thickness of the impurity layer introduced in the experiments and $a_{\rm Al_2O_3}$ is the interatomic spacing in Al_2O_3. From an experimental point of view, if a physical vapor deposition technique is used to prepare these junctions, then this effective thickness $t_{\rm impurity}$ would be determined by the deposition time, the deposition rate having been previously calibrated. We chose $a_{\rm Al_2O_3} = 2.5$ Å in this amorphous oxide. The rest of this submonolayer is made of Al_2O_3 with a concentration $c_{\rm A} = 1 - c_{\rm B}$. The impurities are also characterized by their scattering potentials: $\delta = E_{\rm B}\delta(\boldsymbol{\rho}-\boldsymbol{\rho}_n)\delta(z-0)$ and $\delta^{\rm SF} = E_{\rm B}^{\rm SF}\delta(\boldsymbol{\rho}-\boldsymbol{\rho}_n)\delta(z-0)$ for the spin-conserving and spin-flip scattering respectively. The z-axis is perpendicular to the plane of the structure, the submonolayer of impurities is located at $z = 0$. $\boldsymbol{\rho}$ designates the in-plane coordinates. The impurities are located at the points of coordinates $(\boldsymbol{\rho}_n, 0)$. The tunneling electrons are described in the following way: Let us suppose that the exact wavefunctions propagating from the left side of the barrier $\Psi_{\rm L}^{\sigma}(E_{\rm F}, \mathbf{x})$ and from the right side of the barrier $\Psi_{\rm R}^{\sigma}(E_{\rm F}, \mathbf{x})$ are known. $\sigma = \uparrow, \downarrow$ represents the spin direction, $E_{\rm F}$-Fermi energy and $\mathbf{x}$-the electron's momentum in the plane of the layers. The insulator is described as a flat barrier with height U_0. The imaginary momentum of the electrons inside the barrier is therefore equal to $\mathrm{i}q = \mathrm{i}\sqrt{q_0^2 + x^2}$, with $q_0 = \frac{1}{\hbar}\sqrt{2m(U_0 - E_{\rm F})}$ and m is the effective mass. The system without impurities is then characterized by two parameters which are q and $C^{\sigma}(E_{\rm F}, x)$, the latter being defined by $\mathrm{i}C^{\sigma}(E_{\rm F}, x) \equiv \partial \ln \Psi_{\rm R}^{\sigma}(E_{\rm F}, x, z)/\partial z|_{z=-\frac{b}{2}} = -\partial \ln \Psi_{\rm L}^{\sigma}(E_{\rm F}, x, z)/\partial z|_{z=-\frac{b}{2}}$. In the free electron model, C^{σ} would be defined by $C^{\sigma} = \sqrt{k_{\rm F}^{\sigma 2} - x^2}$ in which $k_{\rm F}^{\sigma}$ is the Fermi-momentum of electrons with spin σ. The free electron model is based on the picture of the band-structure of magnetic transition metals proposed by Stearns [3] and subsequently used by various authors [4,5].

To calculate the two-point conductivity $\sigma(z, z')$, the Kubo formula in mixed real-space(z)/momentum(x) representation is used [5,6]. The bias voltage is supposed to be low enough to stay in the regime of linear response:

$$\sigma(z, z') = \frac{4e^2}{\pi\hbar}\overline{\sum_{x,x',i} A_{xx'}^{\sigma} \overleftrightarrow{\nabla}_z \overleftrightarrow{\nabla}_{z'} A_{xx'}^{\sigma}} \tag{1}$$

where $\overleftrightarrow{\nabla}_z = \frac{1}{2}\left(\overleftarrow{\nabla}_z - \overrightarrow{\nabla}_z\right)$ is the anti-symmetric gradient operator, $A_{xx'}^{\sigma}(z, z') = \frac{1}{2}\left[G_{xx'}^{{\rm ret},\sigma}(z, z') - \left(G_{xx'}^{{\rm ret},\sigma}(z, z')\right)^*\right]$ and $G_{xx'}^{{\rm ret},\sigma}(z, z')$ is the retarded Green function of an electron with spin-index σ. The $\overline{\ }$ symbol denotes the averaging over the distribution of impurities in the submonolayer of impurities. We point out that in the present case, the so-called vertex corrections to the conductivity are not zero, contrarily to the case of macroscopically homogeneous media. This point will be developed further. The Hamiltonian of the considered structure is written:

$$\hat{H} = \hat{H}_0 + \hat{H}_{\rm spin-cons} + \hat{H}_{\rm spin-flip}, \quad \text{with} \tag{2}$$

$$\hat{H}_0 = \sum_{\sigma}\int \mathrm{d}r \Psi^{+\sigma}\left[-\frac{\hbar^2}{2m}\nabla^2 + U^{\sigma}\right]\Psi^{\sigma}(r) \tag{2a}$$

$$\hat{H}_{\rm spin-cons} = \sum_{\sigma,n} \mathrm{d}r\delta(z)\delta(\rho - \rho_n)\varepsilon_n^{\sigma}\Psi^{+\sigma}(r)\Psi^{\sigma}(r) \tag{2b}$$

$$\hat{H}_{\rm spin-flip} = \sum_n \delta(z)\delta(\rho - \rho_n) \times \begin{bmatrix} J_n\left(\Psi^{+\uparrow}(r)\Psi^{\downarrow}(r)S^{-}(\rho_n) + \Psi^{+\downarrow}(r)\Psi^{\uparrow}(r)S^{+}(\rho_n)\right) \\ +\hat{\lambda}_n(\rho_n)\left(\Psi^{+\uparrow}(r)\Psi^{\downarrow}(r) + \Psi^{+\downarrow}(r)\Psi^{\downarrow}(r)\right) \end{bmatrix} \tag{2c}$$

where H_0 describes the free motion of the electrons through the structure, $H_{\rm spin-cons}$ describes the electron scattering on the random potential ε_n of the impurities and $H_{\rm spin-flip}$ describes the spin-flip electron scattering on the spin fluctuations of the impurities (first term in (2c)) and spin-orbit contribution to the scattering (second term in (2c)). $\Psi^{\sigma}(r)$ are field operators, J_n – exchange integral of the electron and impurity spins, $\hat{\lambda}_n$ – operator of the spin orbit interaction. $U^{\sigma} = V^{\sigma}$ for z within the ferromagnetic layers, where V^{σ} represents the spin-dependent position in energy of the bottom of the conduction band in the ferromagnetic layers. $U^{\sigma} = U_0$ for z within the tunnel barrier. ε_n^{σ}, J_n, λ_n take the values $E_{\rm B}^{\sigma}$, $J_{\rm B}$, $\lambda_{\rm B}$ if the impurity is situated on site n and $E_{\rm A}^{\sigma}$, $J_{\rm A}$, $\lambda_{\rm A}$ on the host atom.

In order to calculate the Green functions of Hamiltonian (1), the vertex corrections and finally the conductivity, the coherent potential approximation (C.P.A.) is used [7]. We therefore introduce the coherent potential Σ_{σ} located on the plane of impurities, and the effective Green function $G^{{\rm eff},\sigma,\sigma}$, which is equal in C.P.A. to the exact Green function $G^{\sigma,\sigma}$ averaged over the distribution of impurities:

$$\hat{G}(\rho, z, \rho', z') = \hat{G}^{\rm eff}(\rho, z, \rho', z') + \int G^{\rm eff}(\rho, z, \rho'', 0)\hat{T}G^{\rm eff}(\rho'', 0, \rho', z')\mathrm{d}^2\rho'' \tag{3}$$

where $\hat{T} = \sum_n \hat{T}_n$ is the operator of the full T-matrix, describing the electrons scattering, and $\hat{T}_n$ is the single site T-matrix. A difference from the usual Green function formalism [7] must be pointed out: The Green functions and T-matrix are here matrices in spin space, due to the presence in the Hamiltonian of spin-flip terms. In contrast to [7], $G^{\rm eff}$ describes the motion of the electrons not in a uniform effective field but in a potential which is spatially modulated. However the procedure of the calculation can be easily generalized for the case under consideration. After averaging equation (3) over the configurations of impurities, over the spin S and orbital degrees of freedom,

$$\sigma^{\uparrow}_{\rm AP} = \frac{8e^2}{\hbar\pi^2}\left\{\begin{array}{l}\displaystyle\int_0^{\min\left(k^{\uparrow}_{\rm F1},k^{\downarrow}_{\rm F3}\right)} x{\rm d}x \frac{q^4 c_1^{\uparrow} c_3^{\downarrow} {\rm e}^{-2qb}}{\left(c_1^{\uparrow 2}+q^2\right)\left(c_3^{\downarrow 2}+q^2\right)|2q+\Sigma^{\uparrow}|^2} + \left[\int_0^{k^{\uparrow}_{\rm F1}} x{\rm d}x \frac{q^2 c_1^{\uparrow}{\rm e}^{-qb}}{\left(c_1^{\uparrow 2}+q^2\right)}\right.\\ \left.\times\frac{a_0^2}{2\pi}\left(\displaystyle\int_0^{k^{\downarrow}_{\rm F3}} x{\rm d}x \frac{q^2 c_3^{\downarrow}{\rm e}^{-qb}\Gamma_0^{\uparrow}}{\left(c_3^{\downarrow 2}+q^2\right)|2q+\Sigma^{\uparrow}|^2} + \int_0^{k^{\uparrow}_{\rm F3}} x{\rm d}x \frac{q^2 c_3^{\uparrow}{\rm e}^{-qb}\Gamma_{\rm SF}}{\left(c_3^{\downarrow 2}+q^2\right)|2q+\Sigma^{\downarrow}|^2}\right)\right] + [1\leftrightarrow 3]\end{array}\right\} \quad (10)$$

one obtains:

$$\left\langle \overline{G}\right\rangle = G^{\rm eff} \quad (4)$$

and

$$\left(\begin{array}{cc}\left\langle \overline{T}_n^{\uparrow\uparrow}\right\rangle & \left\langle \overline{T}_n^{\uparrow\downarrow}\right\rangle\\ \left\langle \overline{T}_n^{\downarrow\uparrow}\right\rangle & \left\langle \overline{T}_n^{\downarrow\downarrow}\right\rangle\end{array}\right) = c_{\rm A}\left(\begin{array}{cc}\left\langle \overline{T}_{\rm A}^{\uparrow\uparrow}\right\rangle & \left\langle \overline{T}_{\rm A}^{\uparrow\downarrow}\right\rangle\\ \left\langle \overline{T}_{\rm A}^{\downarrow\uparrow}\right\rangle & \left\langle \overline{T}_{\rm A}^{\downarrow\downarrow}\right\rangle\end{array}\right) + c_{\rm B}\left(\begin{array}{cc}\left\langle \overline{T}_{\rm B}^{\uparrow\uparrow}\right\rangle & \left\langle \overline{T}_{\rm B}^{\uparrow\downarrow}\right\rangle\\ \left\langle \overline{T}_{\rm B}^{\downarrow\uparrow}\right\rangle & \left\langle \overline{T}_{\rm B}^{\downarrow\downarrow}\right\rangle\end{array}\right) = \left(\begin{array}{cc}0&0\\0&0\end{array}\right) \quad (5)$$

where $\langle ...\rangle$ denotes the averaging over spin and orbital degrees of freedom. The explicit expressions for $T_n^{\sigma\sigma'}$ are given by:

$$\left\langle T^{\uparrow\uparrow}_{\rm A(B)}\right\rangle = \frac{\left(E^{\uparrow}_{\rm A(B)}-\Sigma^{\uparrow}_{\rm A(B)}\right)\left(1-\left(E^{\downarrow}_{\rm A(B)}-\Sigma^{\downarrow}\right)F^{\downarrow}\right)+{E^{\rm SF}_{\rm A(B)}}^2 F^{\downarrow}}{Den^1_{\rm A(B)}} \quad (6a)$$

$$\left\langle T^{\downarrow\downarrow}_{\rm A(B)}\right\rangle = \frac{\left(E^{\downarrow}_{\rm A(B)}-\Sigma^{\downarrow}_{\rm A(B)}\right)\left(1-\left(E^{\uparrow}_{\rm A(B)}-\Sigma^{\uparrow}\right)F^{\uparrow}\right)+{E^{\rm SF}_{\rm A(B)}}^2 F^{\uparrow}}{Den^1_{\rm A(B)}} \quad (6b)$$

$$\left\langle T^{\uparrow\downarrow}_{\rm A(B)}\right\rangle = \frac{\left\langle E^{\uparrow\downarrow}_{\rm A(B)}\right\rangle}{Den^{1,\uparrow}_{\rm A(B)}} \equiv 0 \quad (6c)$$

$$\left\langle T^{\downarrow\uparrow}_{\rm A(B)}\right\rangle = \frac{\left\langle E^{\downarrow\uparrow}_{\rm A(B)}\right\rangle}{Den^{1,\downarrow}_{\rm A(B)}} \equiv 0 \quad (6d)$$

$$Den^1_{\rm A(B)} = \left(1-\left(E^{\uparrow}_{\rm A(B)}-\Sigma^{\uparrow}\right)F^{\uparrow}\right)\times\left(1-\left(E^{\downarrow}_{\rm A(B)}-\Sigma^{\downarrow}\right)F^{\downarrow}\right)+\left(E^{\rm SF}_{\rm A(B)}\right)^2 F^{\uparrow}F^{\downarrow} \quad (6e)$$

where we introduced the notation: ${E^{\rm SF}_{\rm A(B)}}^2 = J^2_{\rm A(B)}\langle S^+S^-\rangle + \langle\hat{\lambda}^2_{\rm A(B)}\rangle$, $E^{\uparrow\downarrow}_{\rm A(B)} = J_{\rm A(B)}S^+_{\rm A(B)} + \hat{\lambda}_{\rm A(B)}$, $E^{\downarrow\uparrow}_{\rm A(B)} = J_{\rm A(B)}S^-_{\rm A(B)} + \hat{\lambda}_{\rm A(B)}$, $\langle S^{\alpha}_n S^-_n\rangle$ is the average number of spin fluctuations on site n and $F^{\uparrow(\downarrow)} \equiv G^{\uparrow(\downarrow){\rm eff}}(\rho,0;\rho,0)$. The effective Green function is the solution of the operator equation:

$$\left(E_{\rm F}-\hat{H}^{\rm eff}\right)\hat{G}^{\rm eff}(\rho-\rho',z-z') = \hat{I}\delta(\rho-\rho')\delta(z-z') \quad (7)$$

in which $\hat{H}^{\rm eff}$ coincides with H (expression (2)), where the random potentials ε^{σ}_n are substituted by translationally invariant coherent potentials Σ^{σ}. It is important to notice that due to the identities $\langle S^+\rangle = \langle S^-\rangle = \langle\hat{\gamma}\rangle = 0$, $\Sigma^{\uparrow\downarrow(\downarrow\uparrow)} \equiv 0$ and $H^{\rm eff}_{\rm spin-flip} \equiv 0$ as well. Then, since $H^{\rm eff}$ is translationally invariant in $\boldsymbol{\rho}$-plane, it is easy to solve equation (2) by Fourier transform on the coordinate $\rho-\rho'$. The resulting expression for the function $F^{\sigma} \equiv G^{{\rm eff},\sigma}(\rho,0;\rho,0)$ has the form:

$$F^{\sigma} = -\frac{1}{N}\sum_x \frac{1}{2q+\Sigma^{\sigma}} \quad (8)$$

where for convenience, we expressed the energy in units of $\frac{\hbar^2}{2m}$. The system of equations (5, 6a, 6b, 8) define the unknown coherent potential Σ^{σ}. In the following, its value is determined numerically. To find the vertex corrections, we follow the C.P.A. method, suggested in [8]. We substitute the expression of the exact Green functions (3) into the expression (1) for the conductivity. In the obtained expression, besides the product of two effective Green functions, an additional term appears, proportional to the product of two full T-matrix averaged over impurity configuration and over spin degrees of freedom. This term is called vertex correction $\Gamma(\rho-\rho';z=z'=0) \equiv \Gamma(\rho-\rho')$. Γ is solution of the following integral equation:

$$\begin{aligned}\left(\begin{array}{cc}\Gamma^{\uparrow\uparrow}(\rho-\rho') & \Gamma^{\rm SF}(\rho-\rho')\\ \Gamma^{\rm SF}(\rho-\rho') & \Gamma^{\downarrow\downarrow}(\rho-\rho')\end{array}\right) &= \left(\begin{array}{cc}T^{\uparrow\uparrow} & T^{\rm SF}\\ T^{\rm SF} & T^{\downarrow\downarrow}\end{array}\right)\delta(\rho-\rho')\\ &+\int\left(\begin{array}{cc}T^{\uparrow\uparrow} & T^{\rm SF}\\ T^{\rm SF} & T^{\downarrow\downarrow}\end{array}\right)\\ &\times\left(\begin{array}{cc}G^{{\rm eff}\uparrow 2}(\rho-\rho'')-F^{2\uparrow} & 0\\ 0 & G^{{\rm eff}\downarrow 2}(\rho-\rho')-F^{2\uparrow}\end{array}\right)\\ &\times\left(\begin{array}{cc}\Gamma^{\uparrow\uparrow}(\rho''-\rho') & \Gamma^{\rm SF}(\rho''-\rho')\\ \Gamma^{\rm SF}(\rho''-\rho') & \Gamma^{\downarrow\downarrow}(\rho''-\rho')\end{array}\right){\rm d}^2\rho''. \quad (9)\end{aligned}$$

Due to the translational invariance in the $\boldsymbol{\rho}$-plane of all quantities in equation (9), the latter is easily solved by Fourier transform. After substitution of Γ into the expression of the conductivity, we obtained:

See equation (10) above

where $C_{1(3)}^{\uparrow(\downarrow)} = \sqrt{\left(k_{\mathrm{F1}(3)}^{\uparrow(\downarrow)}\right)^2 - x^2}$. We emphasize again that $k_{\mathrm{F}}^{\sigma} \equiv \partial \ln \Psi_{\mathrm{R}}^{\sigma}(E_{\mathrm{F}}, k_{\parallel}, z)/\partial z|_{z=-\frac{b}{2}}$ in the case of arbitrary band structure of the ferromagnetic layer. Further on, the value of k_F^{σ} will be chosen so as to fit the experimental values of the conductance of the system in absence of impurities. For the free electron model, k_F^{σ} are the spin-dependent radii of Fermi-spheres. The adopted dependence of $C^{\uparrow(\downarrow)}$ on x is justified if the vector $\mathbf{k}_F$ lies near extremum points of the Fermi-surface. Furthermore, the deviation from this dependence is of small importance due to the large value of e^{+2qb}: only small values of in-plane momentum x contributes to the integral over x.
The vertex corrections $\Gamma_0^{\uparrow(\downarrow)}$ and Γ_{SF} are equal to:

$$\Gamma_0^{\uparrow(\downarrow)} = \frac{\overline{(T^{\uparrow(\downarrow)})^2}\left[1-(T^{\uparrow(\downarrow)})^2 D^{\downarrow}\right] + \left(\overline{(T^{\mathrm{SF}})^2}\right)^2 D^{\uparrow(\downarrow)}}{Den^2} \tag{11}$$

$$\text{with} \quad Den^2 \left[1-\overline{(T^{\uparrow})^2}D^{\uparrow}\right]\left[1-\overline{(T^{\uparrow})^2}D^{\downarrow}\right] - \overline{(T^{\mathrm{SF}})^2}D^{\uparrow}D^{\downarrow} \tag{12}$$

where

$$\overline{(T^{\uparrow(\downarrow)})^2} = c_{\mathrm{A}}\left(T_{\mathrm{A}}^{\uparrow(\downarrow)}\right)^2 + c_{\mathrm{B}}\left(T_{\mathrm{B}}^{\uparrow(\downarrow)}\right)^2 \tag{13}$$

$$\Gamma_{\mathrm{SF}} = \frac{\overline{(T_{\mathrm{SF}})^2}}{Den^2} \tag{14}$$

$$\overline{(T_{\mathrm{SF}})^2} = c_{\mathrm{A}}\frac{\left(E_{\mathrm{A}}^{\mathrm{SF}}\right)^2}{\left(Den_{\mathrm{A}}^1\right)^2} + c_{\mathrm{B}}\frac{\left(E_{\mathrm{B}}^{\mathrm{SF}}\right)^2}{\left(Den_{\mathrm{B}}^1\right)^2} \tag{15}$$

and

$$D^{\uparrow(\downarrow)} = \frac{1}{N}\sum_x \left|\frac{1}{2q+\Sigma^{\uparrow(\downarrow)}}\right|^2 - \left|\frac{1}{N}\sum_x \frac{1}{2q+\Sigma^{\uparrow(\downarrow)}}\right|^2. \tag{16}$$

Expression (10) represents the conductance of spin-up electrons in the anti-parallel magnetic configuration. For spin-down electrons and in the parallel magnetic configuration, the expressions for conductance may be obtained from (6) by the appropriate changes in indexes $\uparrow$ and $\downarrow$. The TMR amplitude can then be calculated from: $TMR = \frac{\sigma_p^{\uparrow}+\sigma_p^{\downarrow}-\sigma_{\mathrm{AP}}^{\uparrow}-\sigma_{\mathrm{AP}}^{\downarrow}}{\sigma_{\mathrm{AP}}^{\uparrow}+\sigma_{\mathrm{AP}}^{\downarrow}}$.

From (10–16), it follows that the scattering of tunneling electrons by the impurities introduced in the barrier influences the conductance and therefore the TMR in a twofold way. Firstly, in the denominators of all terms in (6), appears the coherent potential Σ. Consequently, the conductance decreases when the scattering increases *i.e.* when the concentration c_{B} or the scattering amplitude E_{B} increase. The effect of the impurities, which are oxidized together with the metallic aluminum during the formation of the barrier by plasma oxidation [1], is to increase the average height of the potential barrier seen by the tunnel electrons.

Secondly, besides the first term in (10) which represents the direct tunneling [4], there are two additional terms. They describe the tunneling assisted by spin-conserving and spin-flip scattering respectively [9]. These two terms increase the conductance by opening new tunneling channels. Since the spin-conserving contribution is larger for majority electrons in the parallel magnetic configuration ($k_{\mathrm{F1}}^{\uparrow} > k_{\mathrm{F1}}^{\downarrow}$ and therefore $c_1^{\uparrow} > c_1^{\downarrow}$ and same inequalities for layer F3 in (6)), this term increases the value of TMR. In contrast, the spin-flip scattering contribution is larger for minority electrons resulting in a decrease of the TMR amplitude.

It is important to note that the coherent potential Σ increases as a function of the scattering potential beginning as a linear function of the amplitude of spin-conserving scattering potential and concentration c_{B}. In contrast, the spin-flip scattering potential does not contribute to the coherent potential at first order. Meanwhile, the vertex corrections and therefore the "tunneling assisted by scattering" terms in the conductance increase as well with scattering potential but quadratically. Consequently, the total conductance always decreases with increasing spin-conserving scattering potential but may increase with increasing spin-flip scattering potential, since the latter does not give a contribution to the coherent potential at first order.

For comparison with experimental data, the following values of Fermi-wave vectors and scattering potentials were used: $k_{\mathrm{F1}}^{\uparrow} = 1.1$ Å^{-1} and $k_{\mathrm{F1}}^{\downarrow} = 0.5$ Å^{-1} for Co and $k_{\mathrm{F3}}^{\uparrow} = 1.0$ Å^{-1}, $k_{\mathrm{F3}}^{\downarrow} = 0.6$ Å^{-1} for $Ni_{80}Fe_{20}$. Furthermore, we defined $k_{\mathrm{F}}^{\uparrow} = \left(k_{\mathrm{F1}}^{\uparrow} + k_{\mathrm{F3}}^{\uparrow}\right)/2$, $k_{\mathrm{F}}^{\downarrow} = \left(k_{\mathrm{F1}}^{\downarrow} + k_{\mathrm{F3}}^{\downarrow}\right)/2$. For the scattering potentials, the following values were taken: $E_{\mathrm{A}}^{\uparrow} = 0.0$, $E_{\mathrm{A}}^{\downarrow} = 0.0$, $E_{\mathrm{B}}^{\uparrow} = \alpha_1\left[a_0\left(q_0^2 + (k_{\mathrm{F}}^{\uparrow})^2\right)\right]$ and $E_{\mathrm{B}}^{\downarrow} = \alpha_3\left[a_0\left(q_0^2 + (k_{\mathrm{F}}^{\uparrow})^2\right)\right]$ with $\alpha_3 = \alpha_1\left[q_0^2 + (k_{\mathrm{F}}^{\downarrow})^2\right] / \left[q_0^2 + (k_{\mathrm{F}}^{\uparrow})^2\right]$. The used spin-flip scattering potentials were: $E_{\mathrm{A}}^{\mathrm{SF}} = 0.0$ (no spin-flip scattering in Al_2O_3) and $E_{\mathrm{B}}^{\mathrm{SF}} = \gamma E_{\mathrm{B}}^{\uparrow}$.

In the above expressions, α_1 is the scattering potential for up-electrons normalized by the height of the barrier, and γ is the spin-flip scattering potential normalized by the spin-conserving scattering potential for spin-up electrons.

Furthermore the lattice constant in the magnetic layers a_0 is chosen equal to 3.0 Å, the barrier thickness $b =$ 14.0 Å and the evanescent wave-vector in the barrier $q_0 =$ 1.0 Å^{-1}.

In order to compare our calculations to Jansen and Moodera' results [1], the parameters (α_1, γ) were adjusted in our model to fit the experimental results. Figure 1 shows the calculated curves and the experimental points from reference [1] at 77 K and 300 K. The values of the parameters (α_1, γ) used to obtain these fits are listed in Table 1.

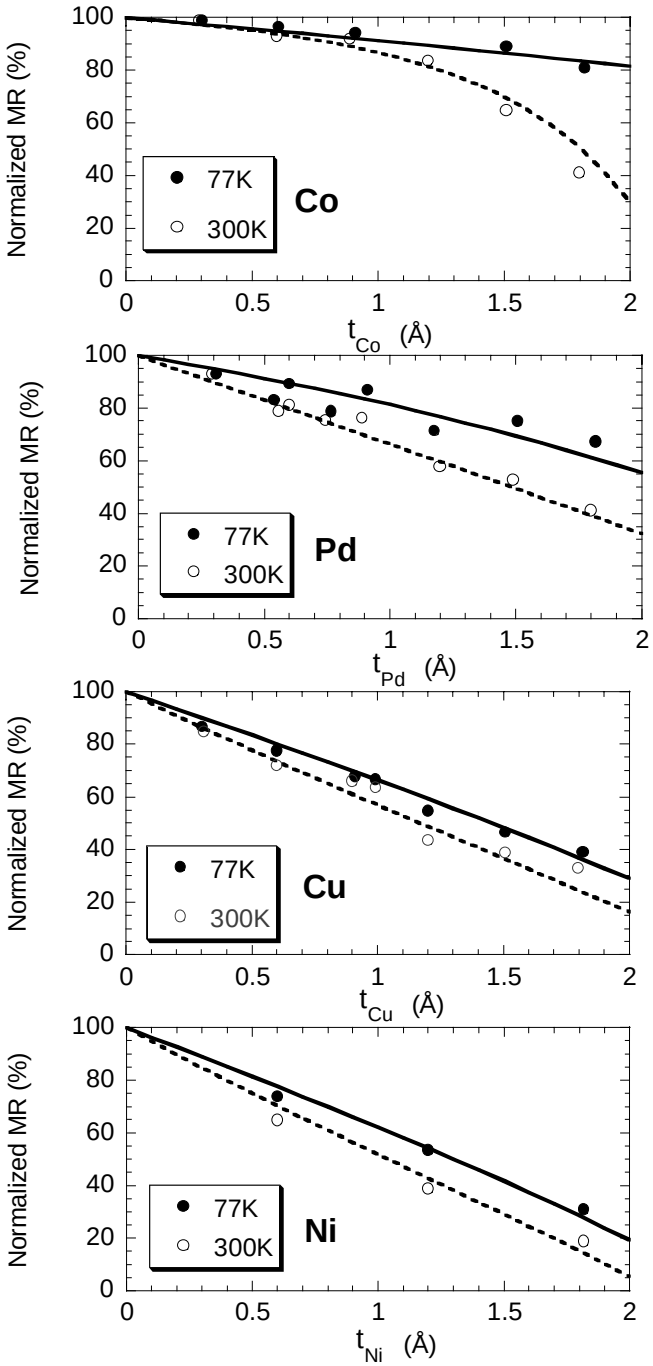

Fig. 1. Normalized magnetoresistance *versus* amount of impurities introduced in the barrier. The points are the experimental data reported from reference [1]. In each plot, the continuous line is the fit of the data at 77 K, the dotted line is the fit for the data at 300 K. Values of the coefficient α_1 and γ are taken from the Table 1.

Very good fits of the experimental data are obtained. In particular, the almost linear decay of magnetoresistance amplitude with the nominal thickness of the impurity submonolayer (except for Co) [1] is well reproduced. For Co, α_1, which characterizes the spin-conserving scattering potential amplitude, varies significantly between 77 K and 300 K whereas for the other elements (Ni, Cu, Pd), no variation is found. To understand the observed temperature dependence one has to take into account that oxides of the impurities introduced in the middle of the barrier may possess intrinsic spins. Moreover it is well known [10] that these oxides may order ferro ($T_{\rm K}$) or antiferromagnetically ($T_{\rm N}$) due to the indirect or superexchange interactions. $T_{\rm K}$ and $T_{\rm N}$ are Curie and Néel temperatures correspondingly. The relatively weak decay of magnetoresistance amplitude *versus* amount of Co impurities was ascribed in [1] to the presence of Co^{3+} ion which has a $3d^6$ electronic configuration and is generally found in a low-spin state [10] with no magnetic moment. However, considering the large variation of α_1 between 77 K and 300 K, a significant fraction of Co^{2+} ion with a corresponding spin of $S = 3/2$ is probably also present. Two contributions to the scattering amplitude α_1 can be expected in these systems: one is the usual Coulomb scattering on impurities, the other is the scattering on fluctuations of the z-component of spin. The former is temperature independent whereas the latter is exponentially small for temperatures much below the ordering temperature ($T_{\rm N}$) and rapidly increases up to the saturation value $S(S+1)$ near $T_{\rm N}$. Since CoO has a Néel temperature very close to room temperature, the large variation in α_1 between 77 K and 300 K can be ascribed to a change in the rate of fluctuation of Co^{2+} spins in this range of temperature.

Table 1. Spin-conserving scattering coefficient α_1 and spin-flip scattering coefficient γ (see text) deduced by adjusting the experimental data of reference [1] to formula (10–16).

Material	Temp = 77 K		Temp = 300 K	
	α_1	γ	α_1	γ
Co	0.03	0.06	0.2	0.1
Pd	0.078	0.1	0.078	0.16
Cu	0.09	0.16	0.09	0.19
Ni	0.103	0.175	0.103	0.206

For the other types of ions, an uncertainty remains regarding the degree of oxidation of the metallic atoms in Al_2O_3 (Cu^+, Cu^{2+} or Cu^{3+}; Ni^{2+} or Ni^{3+}; Pd^{2+}) and their corresponding magnetic properties. However, the very fact that α_1 does not vary between 77 K and 300 K for these ions indicate that the ordering temperature of the spin for these impurities, if any, is either much below 77 K or much above 300 K (for instance with Ni impurities, the Néel temperature in NiO is 520 K).

Concerning the spin-flip scattering amplitude, γ is found to increase for all elements between 77 K and 300 K. Two contributions may be expected. The first one is the spin-wave scattering. Its contribution to $E^{\rm SF}$ and consequently to γ^2 is proportional to $\langle S^+S^-\rangle$ (see definition below (6)). So in spin-wave approximation, $\langle S^+S^-\rangle = \langle S^z\rangle \frac{a_0^2}{2\pi}\int_0^{\pi/a_0} \frac{x{\rm d}x}{{\rm e}^{\frac{D_1x^2+\Delta}{k_0T}}-1} = \beta\frac{T}{T_{\rm k}}\left\{-{\rm Ln}\left[\exp\left(\frac{T_{\rm c}}{T}\right)-1\right]+\frac{T_{\rm c}}{T}\right\}$, for two dimensional (2D) ferromagnetic order and $\langle S^+S^-\rangle = \langle S^z\rangle \frac{a_0^2}{2\pi}\int_0^{\pi/a_0} \frac{x{\rm d}x}{{\rm e}^{\frac{D_2x}{k_0T}}-1} = \left(\frac{T}{T_{\rm N}}\right)^2$ for antiferromagnet, where D_1 and D_2 are spin-wave stiffness constants

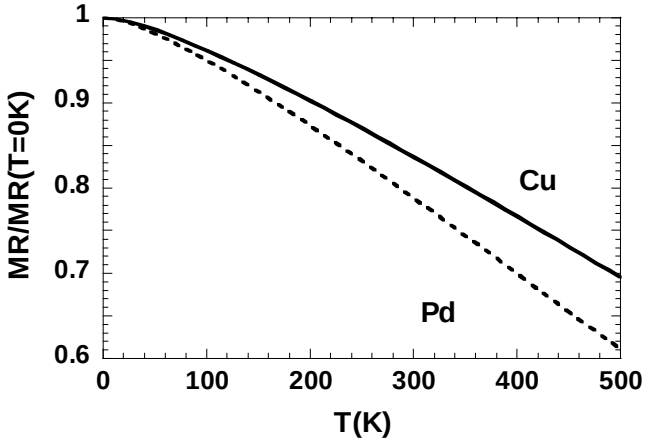

Fig. 2. Calculated variation of magnetoresistance *versus* temperature in junctions doped with Cu or Pd impurities.

and $T_{\rm C} = E_{\rm C}/k_{\rm B}$, $E_{\rm c}$ being the energy gap in the 2D spin-wave spectrum due to the anisotropy at the interface between the electrodes and the insulating layers [11]. The second contribution is due to spin orbit scattering on the impurities. It does not depend on temperature. By summing these two contributions, a phenomenological form of the thermal variation of γ^2 is obtained: $\gamma^2 = \beta \frac{T}{T_{\rm K}} \left\{ -{\rm Ln}\left[\exp\left(\frac{T_{\rm c}}{T}\right) - 1\right] + \frac{T_{\rm c}}{T} \right\} + \gamma_0^2$ for ferromagnet or $\gamma^2 = \beta \left(\frac{T}{T_{\rm N}}\right)^2 + \gamma_0^2$ for antiferromagnet, $\beta = \frac{J_{\rm B}^2}{E_{\rm B}^2}$, $\overline{\gamma}_0^2 = \frac{\langle \lambda_{\rm B}^2 \rangle}{E_{\rm B}^{\uparrow 2}}$. The coefficients β and γ_0^2 can be derived from the values of γ at 77 K and 300 K. From the knowledge of these coefficients and assuming that α_1 does not vary with temperature as found for Cu, Ni, Pd, it is then possible to calculate the expected variation of magnetoresistance as function of temperature in these doped junctions. For illustration, examples of calculated thermal variation of TMR assuming ferromagnetic ordering for Cu and Pd impurities and using the values found for β and γ_0 are shown in Figure 2. A $T_{\rm K} \approx 10$ K was assumed for these impurities. We however point out that the result of the calculation weakly depends on the exact value chosen for $T_{\rm K}$ as long as $T \gg T_{\rm K}$. It would be interesting to measure the thermal variations of magnetoresistance on the same samples used in reference [1] for comparison with these calculated variations.

In conclusion, we presented a quantum mechanical model based on Kubo formalism for the calculation of the magnetoresistance of ferromagnetic tunnel junctions with impurities inside the barrier. The influence of the amount of dopant on the magnetoresistance amplitude was calculated and successfully compared to experiments. From these results, a variation of magnetoresistance *versus* temperature was predicted.

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