Ab-initio calculations of the GMR-effect in Fe/V multilayers

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Abstract

In a self-consistent semi-empirical numerical approach based on ab-initio calculations for small samples, we evaluate the GMR effect for disordered (0 0 1)-(3-Fe/3-V)$_\infty$ multilayers by means of a Kubo formalism. We consider four different types of disorder arrangements: In case (i) and (ii), the disorder consists in the random interchange of some Fe and V atoms, respectively, at interface layers; in case (iii) in the formation of small groups of three substitutional Fe atoms in a V interface layer and a similar V group in a Fe layer at a different interface; and for case (iv) in the substitution of some V atoms in the innermost V layers by Fe. For cases (i) and (ii), depending on the distribution of the impurities, the GMR effect is enhanced or reduced by increasing disorder; in case (iii) the GMR effect is highest, whereas finally, in case (iv), a negative GMR is obtained ('inverse GMR'). © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

The so-called giant magnetoresistance effect (GMR-effect) is well-known; meanwhile, Ref. [1], and several overviews exist on theoretical interpretations, [2–4]. The effect exists in thin film systems made of metallic ferromagnets separated by nonmagnetic or antiferromagnetic metallic spacers,

and consists in the fact that, e.g. in such a trilayer or multilayer system, if one starts from a state, where the ferromagnetic films have mutually antiparallel magnetization, one can switch the magnetization directions, by application of a magnetic field, to parallel orientation, which – as a consequence – generally implies a decrease of the electrical resistance. This amounts typically to values around 10% or so, which is already interesting for applications, but in the case of Fe/Cr/Fe trilayers the effect can be as large as 200% [5].

Thus, the GMR-effect is given by the simple formula

$$\text{GMR} = \frac{\Gamma_{\uparrow\uparrow}}{\Gamma_{\uparrow\downarrow}} - 1, \tag{1}$$

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where the $\Gamma_{\uparrow\uparrow}$ and $\Gamma_{\uparrow\downarrow}$ are the conductances for mutually parallel (resp. antiparallel) magnetizations. Here one distinguishes between the CPP and CIP geometries, where the current is perpendicular to the planes, or in the plane, respectively.

Considering Eq. (1), we would like to stress at this juncture that in a numerical calculation not $\Gamma_{\uparrow\uparrow}$ and $\Gamma_{\uparrow\downarrow}$ separately, but only their *ratio* $\Gamma_{\uparrow\uparrow}/\Gamma_{\uparrow\downarrow}$ must come out correctly.

A considerable GMR effect can already exist in the ballistic regime, i.e. without any impurities, due to the fact that the reflectivity of the electrons at the interfaces changes with the above-mentioned switch. This is shown by the ab initio calculation of Schep et al. for Co/Cu-multilayers in Refs. [6,7], and by recent model calculations of Krompiewski et al. [8,9]. But one of the main problems, namely the question, how impurities and disorder influence the strength and perhaps even the sign of the GMR in a *realistic* system with non-ideal interfaces and significant impurity scattering, is not answered by theories for the ballistic case. For example, it seems almost natural to state that an increase of spin-dependent scattering at the interface should lead to an enhancement of the GMR; on the other hand, one can also imagine that too much disorder at the interface should *reduce* the difference of the spin-dependent reflection properties for the two cases considered above: Thus, without detailed calculations for different systems it remains an open question, whether the GMR is enhanced or reduced by an increasing amount of impurities. Moreover, the answer on this question may depend on the *arrangement* of the impurities.

In the present communication, we study the influence of disorder on the GMR for BCC-(0 0 1)-$(\text{Fe-3/V-3})_\infty$ multilayers, both for CPP and CIP geometry, with four different situations (see below). Our extensive numerical calculations employ a self-consistent semi-empirical approach, which also works for *non-ideal* systems with impurities. We have applied a similar approach earlier for *almost*-ab-initio calculations of the magnetic and transport properties of strongly disordered or even amorphous systems. Both the electronic structure, [10–12], and the transport properties of the systems [12,13] have been calculated, the transport properties directly from the Kubo formula, and separated into

contributions from the up-spin and down-spin carriers. We stress that the Kubo approach is rigorous in principle, and does not invoke the usual Boltzmann approximations. Instead, from the beginning, the disorder of the system is fully taken into account: In particular, in our paper the eigenvalues and eigenvectors of the electronic Hamiltonian are always calculated for the *disordered* system, before the Kubo formula is applied (see below); in the language of diagrammatic theories this means that the vertex corrections are automatically included.

However, all this is possible only for rather small systems: Our computer samples comprise stacks of 12 non-equivalent, partially disordered Fe- or V-monolayers with 4×4 atoms per layer, and with periodic boundary conditions in all three directions. Thus, altogether we have 192 atoms, with 9 orbitals per atom (five 3d-, three 4p-, and one 4s-orbital), i.e. for fixed spin projection $s = \pm 1$ of the electron we diagonalize a Hamiltonian with 1728 lines and columns. After each diagonalization, the expectation values of the local occupations and local moments for every orbital are calculated, and the Hamiltonians are updated, until finally, after a lot of iterations of the procedure, self-consistency is obtained with respect to all local charge and spin expectations (see below). Also the Fermi energies $E_f(s)$ are determined self-consistently. However, in view of the smallness of our systems, the results suffer from the fact that in the numerical calculation we have a discrete spectrum and not a continuum, so that evaluation of histograms instead of continuous functions becomes necessary. (This is different from the technique of Asano et al. [14], which is however not applicable to our system, see below.) Furthermore, one should also be aware of the fact that the $T = 0$ conductance of a mesoscopic physical system is not self-averaging: One needs to average over different samples to obtain significant 'typical results', although with large error bars. But even for these small systems, such results can be obtained, as seen below. This has also been exemplified in former calculations of the resistivity in the magnetic state of disordered systems [12,13].

In the following sections we describe at first our formalism, and then our results for (0 0 1)-(3-Fe/

3-V)$_\infty$-multilayers with four different types of disorder: In cases (i) and (ii), we consider *interchanges* of some nearest-neighbour Fe and V atoms at the interface, whereas in the particular case (iii) we deal with the effect of small Fe 'islands' of three Fe atoms in a V interface layer and vice versa; finally, the case (iv) of a substitutional Fe impurity in an *inner* V layer is considered.

In case (i), we treat the standard situation that the impurities are randomly distributed among all four nonequivalent interfaces, whereas in case (ii) we assume that the impurities are concentrated at only one of the interfaces. Concerning (i), we find at first – as expected – an *increase* of the GMR with increasing impurity concentration, namely from GMR $\sim 60\%$ for an impurity concentration of $n_{imp} \approx 1\%$ to GMR $\sim 120\%$ for $n_{imp} \approx 3\%$ and $\approx 4\%$, whereas in case (ii) a *reduction* of the GMR from $\sim 60\%$ (for $n_{imp} \approx 1\%$) to $\sim 0\%$ (for $n_{imp} \approx 5\%$) is obtained. Also in case (i) the GMR decreases again, if n_{imp} becomes as large as $\approx 5\%$. This different behaviour is discussed below.

For the particular case (iii), see above, we get the largest values of the GMR (~ 250–300%). Finally, in case (iv), the Fe impurity in the inner V layer is significantly magnetized by $-0.5\ \mu_B$, i.e. antiparallel to the Fe layers, if these are aligned in parallel (see below), whereas the Fe impurity is nonmagnetic, if the magnetization directions of the Fe films are mutually antiparallel. In the first-mentioned case the impurity scattering is stronger, which implies that in case (iv) we have a negative GMR ('inverse GMR') of $\sim (-50\%)$. This is true for both the CPP and the CIP geometries, although quantitatively the results are somewhat different for these geometries (see below).

A negative GMR has already been observed in Ref. [15], but for ternary systems, whereas the present system is *binary*.

2. Formalism

We use our realistic self-consistent semi-empirical LCAO approach already described in Refs. [16,12] i.e. with nine orthogonalized orbitals per atom (five 3d-, three 4p- and one 4s-orbital) and a collinear magnetic state described by the equations

$$\sum_{m\beta} H_{l\alpha,m\beta} c_{m\beta,s}^{(v)} + \frac{U_{l\alpha}}{2} \langle n_{l\alpha\uparrow} + n_{l\alpha\downarrow} - 2n_{l\alpha}^{para}\rangle c_{l\alpha,s}^{(v)}$$

$$- \frac{U_{l\alpha}}{2} \langle n_{l\alpha\uparrow} - n_{l\alpha\uparrow}\rangle s c_{l\alpha,s}^{(v)} = \varepsilon_v(s) c_{l\alpha,s}^{(v)}. \qquad (2)$$

In Eq. (2), $c_{l\alpha,s}^{(v)}$ are the probability amplitudes for the event that an electron with spin $s\ (= \pm 1)$ and single-particle energy $\varepsilon_v(s)$ occupies an orbital $\alpha\ (=1,\ldots,9)$ at the site l. Here $v = 1,\ldots,N_l N_\alpha$, where the number of atoms is $N_l = 192$ and the number of orbitals per atom $N_\alpha = 9$. The orbitals are assumed to be orthonormalized, and the real-symmetric matrix $H_{l\alpha,m\beta}$ describes the paramagnetic state of the disordered system. Altogether 26 neighbors, i.e. upto the third shell, are taken into account for each site. The matrix elements are derived from Papaconstantopoulos [17], in the approximation with two-center integrals: We assume that we have a BCC-structure with an averaged lattice constant, $a = (a_{Fe} + a_V)/2$, with $a_{Fe} = 2.87$ Å and $a_V = 3.02$ Å. Thus, it is only necessary to modify the hopping matrices of Papaconstantopoulos according to the modified positions, i.e. for the two-center integrals one uses relations as $I_{dd}(r) \propto r^{-5}$, see Refs. [16,12]; additionally, if site l is occupied by a Fe atom, but site m by V, we assume as usual $H_{l\alpha,m\beta}^{Fe,V} = (H_{l\alpha,m\beta}^{Fe,Fe} H_{l\alpha,m\beta}^{V,V})^{1/2}$. Finally, to agree with *ab initio* calculations, see below, we have used a common shift of $\Delta E = 0.5$ eV for the Fe d-levels with respect to the values in Ref. [17], whereas for V no shift was assumed, and for the Hubbard energies we have taken $U_{l\alpha} = 5.8$ and 2.58 eV for the Fe and V d-orbitals, respectively, $U_{l\alpha} = 0$ otherwise.

Concerning the expectation values in Eq. (2), we require *self-consistency* in the magnetic state, again for the disordered system, namely for every site and every d-orbital we demand that $\langle n_{l\alpha,s}\rangle \equiv \sum_{v=1}^{v_f(s)} |c_{l\alpha,s}^{(v)}|^2$. Here $v_f(s)$ counts the highest occupied single-particle eigenstate for $s = \pm 1$, respectively.

In Fig. 1, the results for the magnetic moments of the different layers for an ideal sample of our system are presented, in comparison with similar results obtained by us for the same system with an ab-initio LMTO method [18]. Obviously, the
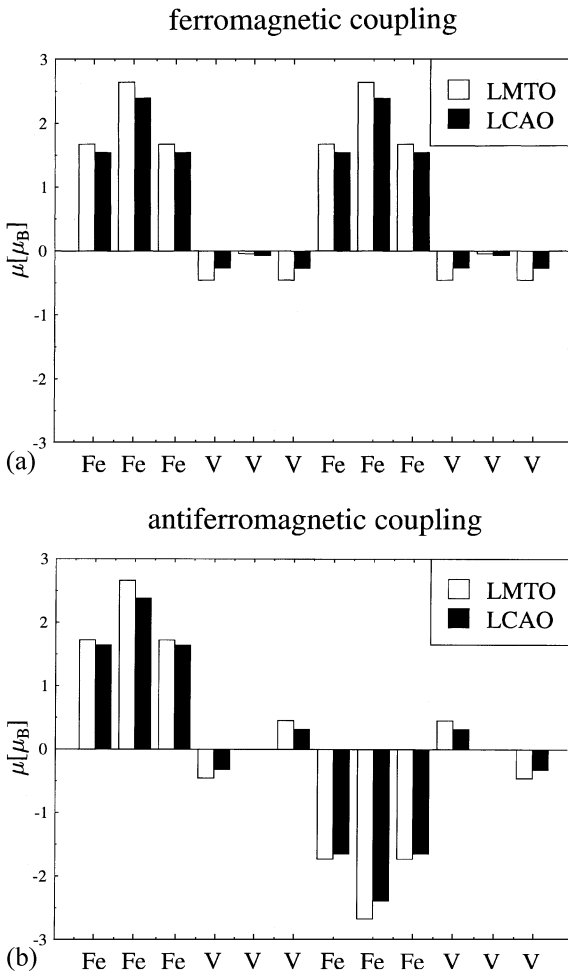
ferromagnetic coupling

antiferromagnetic coupling

Fig. 1. Comparison of the magnetic moments of the pure $(0\,0\,1)$-$(3$-Fe/3-V$)_{\infty}$ multilayers with (a) mutually parallel resp. (b) antiparallel magnetization of the Fe moments, obtained with the ab-initio LMTO program (empty squares) and our self-consistent semi-empirical LCAO approach (full squares). Our elementary cell consists of altogether 12 planes with 16 atoms each, and periodic boundary conditions in x-, y-, and z-direction.

agreement obtained is quite convincing and should give confidence to the reliability of our self-consistent semi-empirical method. Moreover, the following results from Fig. 1 deserve attention: In the central Fe layers, the Fe moments are enhanced to $2.8\,\mu_B$ with respect to the bulk value of $2.2\,\mu_B$, whereas at the interface they are reduced to $1.6\,\mu_B$. On the other hand, vanadium, which is nonmag-

netic in the bulk, has at the interface layers a moment of $(-0.5\,\mu_B)$, i.e. antiferromagnetically coupled to Fe. Already the second V layer, however, is practically nonmagnetic. These results are similar to those obtained by the first-principles LMTO calculations of Süss [19,20].

3. The Kubo formalism

The resistivity is calculated by means of the Kubo formula [21], namely

$$\sigma_{xx}^{(s)} = \frac{e^2\pi}{\hbar\Omega}\overline{(g^{(s)}(E_f))^2(\Delta E(s))^2|\langle f, s|x|i, s\rangle|^2}. \quad (3)$$

Here Ω is the volume of the elementary cell of 192 atoms, $\sigma_{xx}^{(s)}$ the contribution of electrons with spin direction $s = \pm 1$ to the conductivity in x-direction, $g^{(s)}(E_f)$ the value of the spin-dependent density of states of the disordered system at the Fermi energy $E_f(s)$; $|i, s\rangle$ and $|f, s\rangle$ are the exact eigenstates of the full Hamiltonian, again with impurities, with single-particle eigenvalues just above above and below $E_f(s)$, respectively, see below; $\langle \ldots \rangle$ denotes the quantum mechanical expectation value, and the overline denotes an average over 10 samples. $\Delta E(s)$ is a typical energy difference involved in the transition from i to f. To be precise, we choose $\Delta E(s) = \varepsilon^{v_f+1}(s) - \varepsilon^{v_f-1}(s)$, if the highest occupied single-particle eigenstate of spin s has single-particle energy $\varepsilon^{v_f}(s)$. Furthermore, although the eigenstates of our Hamiltonian have been calculated with periodic boundary conditions in x, y, and z-direction, we assume in Eq. (3), for the calculation of the resistivity, that the resistance is measured with parallel planar contacts of a distance as small as our cluster sizes $\Delta x = \Delta y = 4a = 11.78$ Å and $\Delta z = 12\ (a/2) = 17.67$ Å, respectively. Thus, even without impurities we assume an inelastic dephasing-length of this short size, i.e. by a factor $\frac{2}{3}$ shorter in x- and y-directions than for the z-direction. As a consequence of this factor $\frac{2}{3}$, our in-plane conductivities should be scaled by a factor $(\frac{3}{2})^2 = 2.25$, if a direct comparison with the *perpendicular* conductivity is desired. In any case, these correction factors do not enter the GMR, since the ratio $\Gamma_{\uparrow\uparrow}/\Gamma_{\uparrow\downarrow}$ in Eq. (1) does not depend on them.

Concerning the average of Eq. (3), for $|i, s\rangle$ and $|f, s\rangle$ we take the $(n + 1)$ highest occupied and n lowest unoccupied states, respectively, for given s, with $n = 1, \ldots, 5$ in Fig. 2; $n = 1$ otherwise. Additionally, as usual with the Kubo formalism for DC-conductivity, although it would be rigorous only in the thermodynamic limit, we also include the case $i = f$ in Eq. (3), so that the average in this equation is dominated by the $2n + 1$ diagonal terms. Finally, the origin of our coordinates is fixed in such a way that the matrix element $\langle i, s|x_k|i, s\rangle$ would give the actual length of our elementary cell in k-direction ($k = x, y, z$), i.e. the distance of the contacts, for constant $|i, s\rangle$.

4. Results for the GMR

4.1. Pure sample, cases (i) and (ii), and some remarks

In Fig. 2, which only should be considered as a check of the accuracy of our method and also serves for the statement (see below) that without impurities we obtain GMR ≈ 0, we present our (fictitious) results for the CPP- (resp. CIP-) conductivities of pure samples obtained with the different dephasing lengths of $\Delta z = 17.67$ Å and $\Delta x = \Delta y = 11.78$ Å, corresponding to our sample size, see above. Taking the average over the five cases of $(2n + 1)$, one gets the dotted lines, from which one concludes $\rho_{CIP}/\rho_{CPP} \sim 2$, as expected from $(\Delta z/\Delta x)^2 = 2.25$. The error bars of our results amount to $\sim \pm 20\%$. Since these results apply both to the cases of mutually antiparallel (resp. parallel) magnetizations of the Fe films, we conclude that in our case the *GMR-effect in the pure systems vanishes within our accuracy*. For the disordered systems this will be different, see below.

Additionally, at this place the following remarks are in order:

(1) It is not important that due to our short dephasing length our values for ρ_{CPP} and ρ_{CIP} are ~ 2 (resp. ~ 4) times larger than the 'experimental estimates', which one would expect at room temperature [22,23]. Since these rescaling factors of ~ 2 (resp. 4) do not depend on whether one considers $\Gamma^{\uparrow\uparrow}$ or $\Gamma^{\uparrow\downarrow}$, the GMR itself should be insensitive against these rescalings, as already mentioned in connection with Eq. (1).

(2) This is supported by the observation that the numerical results for the resistivities in the magnetic state of disordered Fe/Ni/Mn alloys in our former paper [12] have also come out too large just by a constant factor ~ 5, in spite of the fact that with increasing Mn concentration the conductance decreased considerably. However again, apart from the constant factor, the *concentration dependence* of
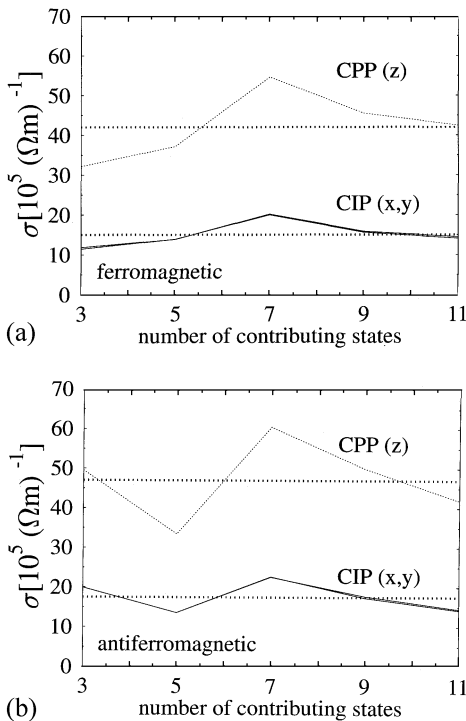

Fig. 2. Conductivities of the pure (001)-(3-Fe/3-V)$_\infty$ multilayers, with contacts as described in the text. 'Ferromagnetic' (resp. 'antiferromagnetic') means that the magnetization directions of the Fe layers are mutually parallel resp. antiparallel. The number $(2n + 1)$ of contributing states for given s, in the vicinity of the spin-dependent Fermi-energy $E_f(s)$, ranges from 3 to 11. The dotted lines represent the averages of the five cases of $(2n + 1)$. The apparent difference between the CPP and CIP conductivities ('current perpendicular to the planes' and 'current in the planes') is not realistic: It results simply from the different distances of the contacts for CIP and CPP, and can be scaled away (see the text). Finally, within our accuracy, the GMR is zero for the present pure system, i.e. the results for the different mutual magnetizations are practically identical. For further details see the text.

the experimental values was well-reproduced in Ref. [12] with our formalism.

(3) Asano et al. [14] have performed a schematic model calculation for pure and impure systems with only s-bands, only nearest-neighbor hopping in simple-cubic arrays, and where the magnetism was not treated self-consistently, but replaced by a spin-dependent constant potential $V_\uparrow = -0.5$, $V_\downarrow = +0.5$ for the magnetic atoms, whereas for the nonmagnetic atoms, $V \equiv +0.5$ was chosen. Finally, even the Fermi energy was arbitrarily fixed at $E_f = 0$. (Here the magnitude of the nearest-neighbor hopping integral has been used as energy unit.) Due to these simplifications, Asano et al. could use a *recursive technique*, where planes with 12×12 atoms could be treated, and 'perfect leads' could be attached to the sample in the current direction. Although within our group this powerful technique has already been extended to extremely accurate model calculations of the CPP-GMR and of a corresponding Giant Magneto-Thermopower in pure s-band tight-binding samples with infinite planes [8], the method is not applicable to the present system, since s-, p-, and d-bands, self-consistency, and interactions upto third-nearest neighbors, are needed. However, at the end we will discuss our results in the light of Ref. [14].

After these preparations we now consider the impure systems: In Fig. 3, we present the results for the GMR obtained with Eqs. (3) and (1) for case (i), i.e. impurities distributed randomly across the four interfaces (see below). The concentrations considered correspond to 1, 2, …, 5 interchanged impurity pairs, e.g. V impurities in a Fe plane and vice versa. Since the volume corresponds to 192 sites, and since to every V impurity position in a Fe plane there is a neighboring Fe impurity in the adjacent V plane, the impurity concentrations range from 1.04 to 5.2%, and we have produced our random samples in such a way that for $n_{imp} \leqslant 4.16\%$ the impurity pairs are situated at *different* interfaces, whereas for $n_{imp} = 5.2\%$ (resp. 6.24%) at one of the four interfaces (resp. two of them) two pairs are situated. The error bars in Fig. 3 result from the average over 10 samples, and the evaluation of Eq. (3) has been performed as in Fig. 2, but with $(2n + 1) = 3$, separately for $s = \pm 1$.
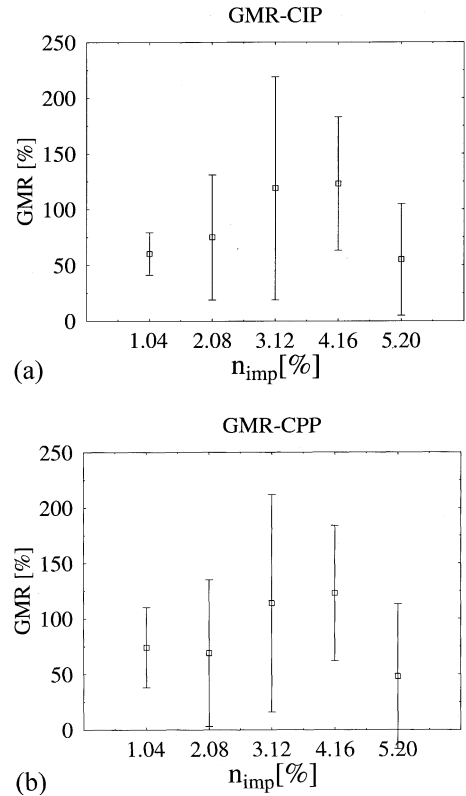

Fig. 3. CIP- and CPP-GMR (see Eq. (1)) for impurity distributions of class (i). This class corresponds to 'interchange impurities' concentrated at upto four *different* interfaces, n with 16 sites per plane. For concentrations of upto 4.16%, every (Fe–V) pair of interchanged atoms has its own interface, whereas in case of 5.2% impurities, at one of four interfaces there are not one, but two of such (Fe–V) pairs. Note that for this class the GMR is mainly *increasing* with the impurity concentration.

It is essential that in spite of the large error bars there is a clear trend in the concentration dependence in Fig. 3, namely at first a roughly linear *increase* from GMR ~ 60% at $n_{imp} \approx 1\%$ to GMR ~ 120% at $n_{imp} \approx 4\%$, which is then followed by a *decrease* to GMR ~ 50% for $n_{imp} \approx 5\%$. (Almost the same results, GMR = 32 and 53% for the CIP and CPP cases, respectively, are obtained with 6% impurities.) The small difference of the CPP-GMR with respect to the CIP-GMR is insignificant; what is of only importance is that the behavior of the GMR with increasing concentration is the same for both cases.

The increase observed upto $n_{imp} = 4.2\%$ is what one would expect by an incoherent superposition of the effects from single impurities. However, for $n_{imp} = 5.24\%$, as already mentioned, one of the interfaces must host two impurity pairs. That this fact leads to a decrease of the GMR, is in agreement with the behavior in case (ii), which is presented in Fig. 4: In this case, where – as discussed above – the impurities are concentrated at only one of the four interfaces, the GMR decreases from $\sim 60\%$ for $\approx 1\%$ impurities down to GMR $\sim 0\%$ at $n_{imp} \approx 5\%$.

The different behavior of cases (i) and (ii) is not easily understood, since it involves the ratio $\Gamma^{\uparrow\uparrow}/\Gamma^{\uparrow\downarrow}$ of two conductances: According to details of our results, which we do not present as plots [24], both $\Gamma^{\uparrow\uparrow}$ and $\Gamma^{\uparrow\downarrow}$ decrease significantly with increasing n_{imp}. Concerning $\Gamma^{\uparrow\downarrow}$, we find that this decrease is roughly the same for the cases (i) and (ii), respectively: E.g. $\sigma_{CIP}^{\uparrow\downarrow}$ decreases from $\sim 11 \times 10^{-5}$ $(\Omega\,cm)^{-1}$ at $n_{imp} \approx 1\%$ to $\sim 6 \times 10^{-5}$ $(\Omega\,cm)^{-1}$ at $n_{imp} \approx 5\%$, both for (i) and (ii), in spite of the different spatial impurity distributions of these cases. In contrast, $\Gamma^{\uparrow\uparrow}$ is found to be quite *sensitive* to the spatial distribution of the impurities and thus essentially responsible for the different behavior of the GMR: In case (i) the decay is rather weak, e.g. $\sigma_{CIP}^{\uparrow\uparrow}$ decays in a 'sub-Boltzmannian way', namely $\propto (7.67 + 9.33 n_{nimp}^{-1})$, from 17×10^5 $(\Omega\,cm)^{-1}$ at $n_{imp} \approx 1\%$ to 10×10^5 $(\Omega\,cm)^{-1}$ at $n_{imp} \approx 4\%$, whereas in case (ii), where the impurity pairs are randomly concentrated at one of our four interfaces, the decay is much faster, namely from 17×10^5 $(\Omega\,cm)^{-1}$ down to 5×10^5 $(\Omega\,cm)^{-1}$, and essentially 'non-Boltzmannian', namely linear in n_{imp}^{+1} instead of n_{imp}^{-1}. This means that vertex corrections and multiple scattering could play an essential role for $\Gamma^{\uparrow\uparrow}$ in case (ii), which is not unreasonable in view of the essentially two-dimensional nature of the scattering for that case, and the erratic magnetization profiles obtained in Fig. 6.

4.2. Magnetic-moment profiles

In Figs. 5 and 6 we present the distribution of magnetic moments in disordered Fe and V interface planes with one (resp. five) (non-neighboring) interdiffusions at the same interface. As one can see from these plots, there is a significant reduction of the Fe moments in the vicinity of the V impurity. Also the V impurities in the Fe planes have a considerable magnetic moment of $\sim -0.8\,\mu_B$, much higher than in the pure V interface plane, where the V moment is only $\sim -0.25\,\mu_B$. This agrees with first-principles calculations of Coehoorn [25].

4.3. Cases (iii) and (iv)

We get a much larger GMR effect than that obtained with cases (i) and (ii) by replacing the *interchange process* of Fe and V neighbors at the interface by the following particular *'island substitution'* process (iii): We substitute randomly three
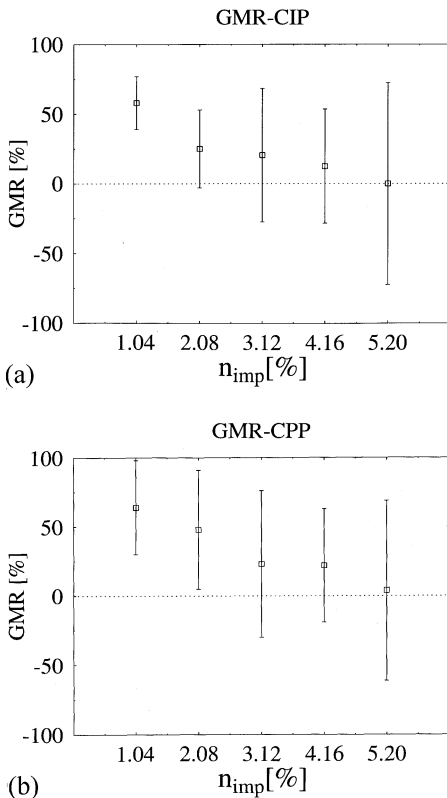

Fig. 4. Same as Fig. 3, however for impurity distributions of class (ii). This class corresponds to 'interchange impurities' concentrated at only one of four interfaces with 16 sites per plane, i.e. for (Fe–V)-pairs at the 'impure interface', the Fe-atoms are substituted by V and vice versa. Note that for this class the GMR is decreasing with increasing impurity concentration.

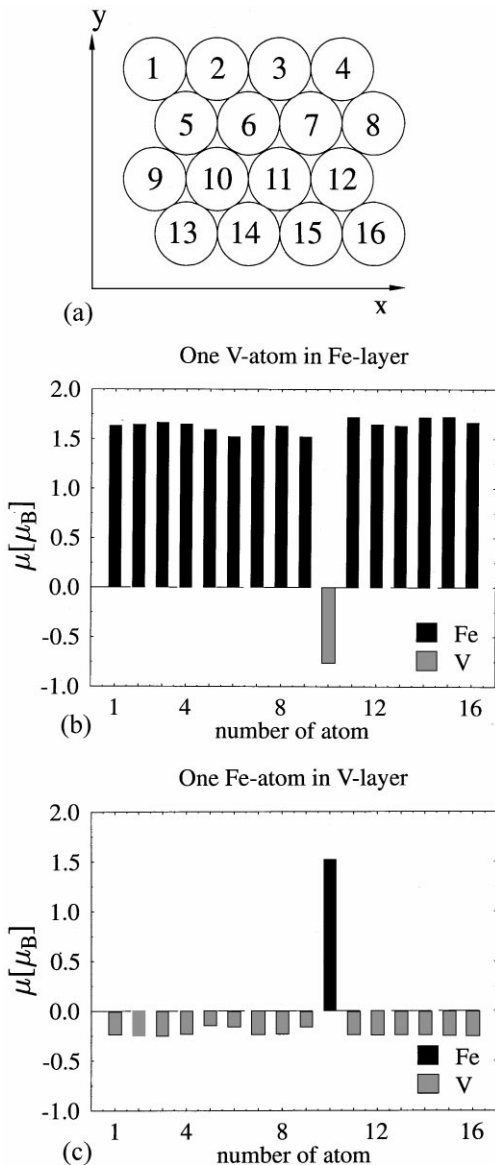Fig. 6. Same as in Fig. 5, however for interface planes with five impurities (out of 16 sites).

Fig. 5. The spatial distribution of the magnetic moments is presented for a Fe interface plane with one V impurity (b) and for a V interface plane with one Fe impurity (c). The relation between the *numbers* 1, 2, ..., 16 given to the atoms and their *positions* in space follows from (a).

neighbouring V atoms in a V interface layer by Fe, and at a different interface in a Fe layer independently three neighboring Fe atoms by V, which corresponds to ∼3.12% impurities. Of course,

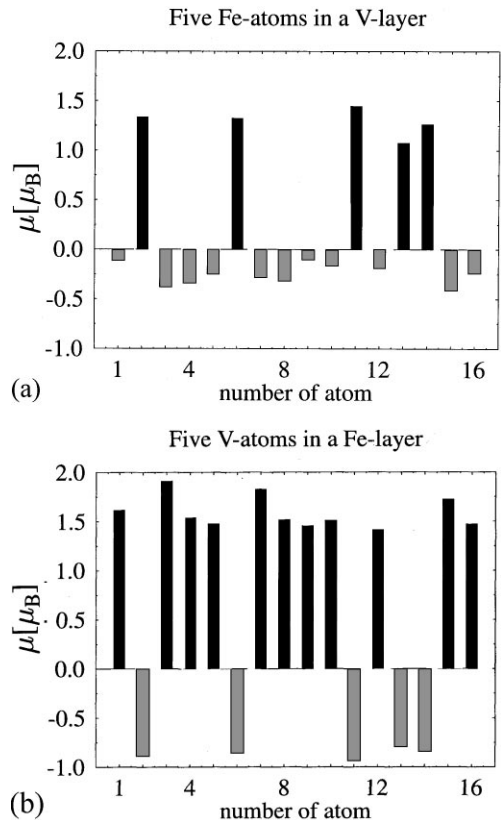there are many possibilities of such a simultaneous random substitution of a small 'island' of three neighboring Fe interface atoms by V and three neighboring V atoms at a different interface by Fe, e.g. atoms 1, 2, 3 or 2, 5, 6 or 2, 6, 10 or ... in Fig. 5a. Averaging over 10 random samples, we obtain a CIP-GMR as large as $(265 \pm 120)\%$ and a CPP-GMR as large as $(300 \pm 170)\%$. These high values with large scatter, which should be contrasted to GMR ∼ 120% obtained for $n_{imp} \approx 3\%$ in case (i), are not yet understood at present, however it is clear that the differences point again to 'non-Boltzmannian' behavior and the possible role of vertex corrections. Probably, it is important that the interfaces become more 'diffuse' for 'interchange impurities' of type (i) and (ii), whereas in case (iii), although the width of the Fe films (V films) varies

locally, the interface remains well-defined in a sense.

Finally, in case (iv), we discuss the situation that there is just one Fe substitutional impurity (i.e. a concentration of 0.5%) in one of the two *central* V planes of our sample. In this case, if adjacent Fe films are magnetized in opposite direction, the Fe moment at the central V layer vanishes on symmetry grounds; but if the Fe films are aligned in parallel, there is a considerable moment induced at the Fe impurity.

In Fig. 7 we present the magnetization profile in the relevant central V layer for this case of mutually parallel magnetic polarizations of the adjacent Fe films. As already mentioned for the pure system, the V polarization in the central plane is almost negligibly small, but the Fe impurity moment is not; instead, it is magnetized antiparallel to the adjacent Fe films, with $\sim 0.5\,\mu_B$, whereas in the Fe films themselves one has the results of Fig. 1, namely $\mu \sim 1.5$ and $2.5\,\mu_B$ at the interfacial (resp. central) Fe layers. From this large induced negative polarization, one can imagine that the present Fe impurity in 'bulk' V induces strong scattering effects in the case of mutually *parallel* polarization of the Fe films, which is unusual, since now one expects a *negative* GMR. Averaging with respect to the few different possibilities to place the impurity with respect to the contacts in Eq. (3), we get in fact (upto $\sim 20\%$ accuracy) the following results: CIP-GMR $\sim 47\%$ and CPP-GMR $\sim -61\%$.

4.4. Discussion

Although a direct comparison is not possible in view of the differences discussed above, we discuss our results in the light of the paper [14] of Asano et al. As already mentioned, these authors treat a very simplified s-band model only, but for larger systems, with attached ideal leads, and with a powerful recursion method.

At first we stress that the values for the CPP-GMR in Ref. [14] are of the same order as ours in case (i), namely $\sim 45\%$ (compared to our 60%) for $n_{imp} \approx 1\%$. This corresponds to the 'interface roughness' $\lambda \approx 0.06$ in Ref. [14], although these authors do not generate the impurities by pairwise interchange, but by simple substitutions in interface
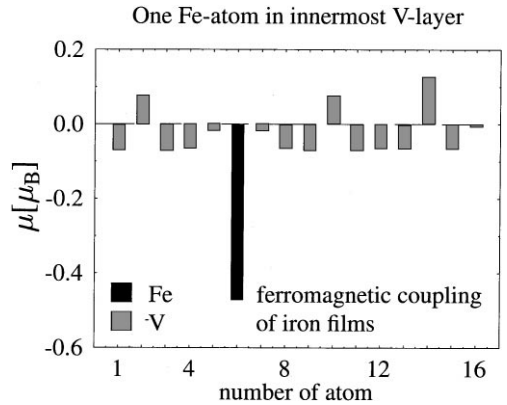

Fig. 7. Same as in Fig. 5, but for case (iv), i.e. there is now only one Fe impurity in a *central* V plane. The Fe films are magnetized in parallel ('ferromagnetic coupling'). In this case there results a negative GMR, see the text.

layers, which has a less drastic effect on the local moments, see Ref. [26]. But with increasing interface roughness, in Ref. [14], the CPP-GMR only *decreases* rather slowly, e.g. down to $\sim 35\%$ at $\lambda \approx 0.3$, while the CIP-GMR, which vanishes for the pure system, *increases* still more slowly. Only, if at all sites i the potentials $V_s(i)$ are additionally randomized by addition of terms $\delta V_s(i)$, which are uniformly and independently distributed between $\pm W_B/2$; Asano et al. obtain a more drastic decrease of both GMRs with increasing W_B, which becomes very rapid for $W_B \gtrsim 1$.

Thus, there are two main differences from our results: (a) Whereas in our case the CIP-GMR is only slightly lower than the CPP-GMR (see above), in Ref. [14] it always remains significantly smaller, e.g. by a factor $\lesssim 0.25$ for $\lambda \approx 0.2$. (b) Concerning the CPP-GMR, according to Ref. [14], for the pure samples it is even somewhat higher than with impurities, whereas in our case it practically vanishes for pure samples (see above). These differences are due to the fact that in Ref. [14] the sample is attached to perfect leads, i.e. the dephasing length is ∞ and the energy spectrum continuous, and the extension of the samples in *x*- and *y*-direction is rather large, whereas in our case we assume periodic boundary conditions, but an inelastic dephasing length as short as our sample sizes, and similarly small widths Δx and Δy.

Therefore, our case $n_{imp} = 0$ does not correspond to the usual 'ballistic situation', in contrast to that in Ref. [14]. However, for our disordered systems the elastic scattering lengths are as short as (or even shorter than) the inelastic dephasing length, and for the GMR of our small and essentially cubic samples, the distinction between 'longitudinal' and 'perpendicular' may become blurred in the presence of impurities and short inelastic dephasing.

The essential point however seems to be the following. According to Ref. [14], disorder and impurities apparently always *reduce* the CPP-GMR, whereas for our small systems with the strong 'magnetic contrast' of Fe and V and the presence of strongly different local situations, also an *increase* can occur, and even a *negative* GMR may be possible. Actually however, in cases (i) and (ii), if there are two or more impurities at an interface plane, i.e. for $n_{imp} \geqslant 1.04\%$ in case (ii), or for $n_{imp} \geqslant 5.2\%$ in case (i), also in our results the GMR always *decreases* with increasing disorder.

Experimentally, it has been found in Ref. [27] that increasing the interface roughness by Xe^+ irradiation leads to a significant enhancement of the GMR; however, beyond a certain irradiation dose the GMR decreased again, which would fit to our main scenario (i).

5. Conclusions

We have applied a self-consistent semi-empirical *almost* ab-initio approach to small samples to calculate the magnetic properties and the CIP- and CPP-GMR effects for $(0\,0\,1)$-$(3\text{-Fe}/3\text{-V})_\infty$ multilayers, with impurities generated by randomly interchanging neighboring Fe and V atoms (i) at all four interfaces, and (ii) only one of the interfaces, for impurity concentrations ranging from 1 to 5% (sometimes 6%). In case (i) we observed an increase of the GMR from $\sim 60\%$ for 1% to $\sim 120\%$ at 4% impurities, followed by a decrease back to $\sim 50\%$ for 5 and 6% impurities, whereas in case (ii), we have found a decrease of the GMR from $\sim 60\%$ for 1% to ~ 0 at 5% impurities. Still much larger GMR values of ~ 250 to $\sim 300\%$ have been obtained in case (iii) for impure systems with small 'islands' of three randomly chosen neighboring Fe

substitutions in a V interface layer and simultaneous, but independent V substitutions in a Fe layer at a different interface, which corresponds to $\approx 3\%$ impurities. Finally, in case (iv), we considered Fe impurities in 'bulk' V, by replacing one of 16 V atoms in one of two central V layers by Fe. This leads in our calculation to a negative GMR of $\sim (-50\%)$. It is remarkable that here a negative ($=$'inverse') GMR has been obtained in a binary system, whereas hitherto this was observed only in ternary systems [15].

Of course, our results should be taken with care: One should be aware of the smallness of the systems (192 atoms), the small 'dephasing distances' between the voltage contacts ($\Delta x = \Delta y \approx 12$ Å, $\Delta z \approx 18$ Å), the large number of strongly reflecting Fe/V-interfaces (two of three monolayers are interface layers), the small concentrations of strongly scattering impurities involved (upto 5% only, Fe impurities in V and vice versa), and the large error bars of the sample averages. However, in spite of these *caveats*, the results seem to justify the statement that the influence of magnetic impurity scattering on the GMR may be more complicated than expected.

Acknowledgements

References

[1] M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Frederich, J. Chazelas, Phys. Rev. Lett. 61 (1988) 2472.

[2] P.M. Levy, Shufeng Zhang, J. Magn. Magn. Mater. 164 (1996) 284.

[3] I. Mertig, P. Zahn, M. Richter, H. Eschrig, R. Zeller, P. Dederichs, J. Magn. Magn. Mater. 151 (1995) 363.

[4] P. Zahn, I. Mertig, M. Richter, H. Eschrig, Phys. Rev. Lett. 75 (1995) 2996.

[5] G. Binasch, P. Grünberg, F. Saurebach, W. Zinn, Phys. Rev. B 39 (1989) 4828.

[6] K.M. Schep, P.J. Kelly, G.E.W. Bauer, Phys. Rev. Lett. 74 (1995) 586.

 [7] G.E.W. Bauer, K.M. Schep, P.J. Kelly, J. Magn. Magn. Mater. 151 (1995) 369.

 [8] S. Krompiewski, U. Krey, Phys. Rev. B 54 (1996) 11961.

 [9] S. Krompiewski, M. Zwierzycki, U. Krey, J. Phys. CM 9 (1997) 7135.

[10] U. Krey, S. Krompiewski, U. Krauss, J. Magn. Magn. Mater. 86 (1990) 85.

[11] U. Krey, U. Krauss, S. Krompiewski, J. Magn. Magn. Mater. 103 (1992) 37.

[12] A. Paintner, F. Süss, U. Krey, J. Magn. Magn. Mater. 154 (1996) 107.

[13] U. Krey, U. Krauss, S. Krompiewski, T. Stobiecki, J. Magn. Magn. Mater. 116 (1992) L7.

[14] Y. Asano, A. Oguri, S. Maekawa, Phys. Rev. B 48 (1993) 6192.

[15] J. Binder, P. Zahn, I. Mertig, J. Magn. Magn. Mater. 100 (1997) 100.

[16] F. Süss, U. Krey, J. Magn. Magn. Mater. 125 (1993) 351.

[17] D.A. Papaconstantopoulos, Handbook of the Bandstructure of Elemental Solids, Plenum Press, New York, 1986.

[18] O.K. Andersen, O. Jepsen, D. Glötzel, in: F. Bassani, F. Fumi, M.P. Tosi (Eds.), Highlights in Condensed Matter Theory, North-Holland, New York, 1985, p. 11. We would like to thank Prof. O.K. Anderson for the possibility to use his LMTO package.

[19] F. Süss, U. Krey, S. Krompiewski, Acta Phys. Pol. A 91 (1997) 281.

[20] F. Süss, Ph.D. Thesis, Regensburg, 1996.

[21] N.F. Mott, E.A. Davis, Electronic Processes in Non-crystalline Materials, Clarendon Press, Oxford, 1979.

[22] The expected values are $\rho_{CPP} \approx (\rho_{Fe} + \rho_V)/2 \approx 14\ \mu\Omega$ cm and $\rho_{CIP} \approx 2\rho_V\rho_{Fe}/(\rho_{Fe} + \rho_V) \approx 12\ \Omega$ cm, obtained with $\rho_{Fe} = 8.6\ \mu\Omega$ cm and $\rho_V = 19\ \mu\Omega$ cm [23] and the values calculated from Fig. 2 are $\rho_{CPP} \approx 24\ \Omega$ cm and $\rho_{CIP} \approx \mu\Omega$ cm.

[23] J. Bass, in: Landolt-Börnstein (Ed.), Numerical Data and Functional Relationships in Science and Technology, vol. 15a, Springer, Berlin, New York, 1982, p. 1.

[24] A. Moser, Diploma Thesis, University of Regensburg, 1991, unpublished.

[25] R. Coehoorn, J. Magn. Magn. Mater. 151 (1995) 341.

[26] J. Pirnay, U. Krey, S. Krompiewski, J. Magn. Magn. Mater. 93 (1991) 267.

[27] D.M. Kelly, I. Schuller, V. Korenivski, K.V. Rao, K.K. Larsen, J. Bottiger, E.M. Gyorgy, R.B. Van Dover, Phys. Rev. B 50 (1994) 3481.