Mössbauer study and Monte Carlo simulations of the hyperfine field distribution in Magnetic/Non-Magnetic (M/NM) multilayers

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Abstract: Using a Monte Carlo numerical method, we perform a complementary analysis of the hyperfine field distribution in Magnetic/Non-Magnetic (*M=Fe/NM=Cu)* multilayers and a comparison with Mössbauer experiment results of $[Fe(40)=Cu(20)]_{20}$ multilayers. From differing relaxation rates for spins with few near neighbor spins and particularly those located at the interface, the Mössbauer result can be simulated. Our model consists of an alternate stacking of magnetic and non-magnetic layers (M_{nM}/NM_{nNM}) with disordered interface. The simulation results confirm that the concentration of interface alloys ($M_{1-x}NM_x$) and magnetic layer thickness modify systemically the magnetization distribution in the *M/NM* multilayers. The result is in agreement with Mössbauer experimental analysis.
PACS: 87.55.K-, 75.70.-i, 67.30.er, 67.30.hp, 75.10.Jm, 75.70.Cn, 32.10.Dk, 36.40.Cg

Keywords: Monte Carlo simulation, Heisenberg model, Multilayers, Interface Morphology, Curie temperature, Mössbauer spectroscopy.

I. Introduction

Multilayer structures constructed by alternating layers of ferromagnetic materials such as *Co*, *Fe* and *Ni*, and separated by «non-magnetic» materials (Ag, Au, Cu ...) are very attractive since they may exhibit in some conditions interesting magnetic properties: the novel magnetic behaviours which have only been observed for films with layers thicknesses on the order of several atomic layers, as well as the possibility of fabricating new materials for practical applications as materials with high giant magneto-resistance (*GMR*) [1-3].

The magnetic properties of *M/NM* multilayers depend highly on the thickness and nature of the interface between layers. Also, the growth conditions introduce changes in the crystal structure of these multilayers and make the study of the magnetic proprieties very difficult. A more detailed interpretation of the effects of interface morphology is required. In particular, the dynamic nature of the spin system must be considered in order to give an explanation of the evolution of the observed Mössbauer spectra with temperature and structural parameters.

In this work, we present a Monte Carlo (MC) investigation of the magnetic properties of *M/NM* multilayers consisting in classical Heisenberg spins. The magnetic thickness (n_M) in *M/NM* multilayers was varied systemically. Our goal is to provide a numerical model reproducing and explaining experimental results on *(M/NM=Fe/Cu)* multilayers and developing a fundamental understanding of the structure and morphology dependence of the magnetic properties.

II. Experimental result

The Mössbauer spectra of *M/NM=* $[Fe(40Å)/Cu(20Å)]_{20}$ multilayers with $M_{Fe}=2.2\mu_B$ collected at room temperature is shown in Figure 1; The spectrum is the superposition of two components: a mean component with hyperfine parameters are those of bcc iron located in the centre of the layers of iron and broad component representing iron atoms involved at the interface [4]. The hyperfine field distribution relative to the broad component has been calculated using the combined hyperfine fields and isomer shift (Figure 2).

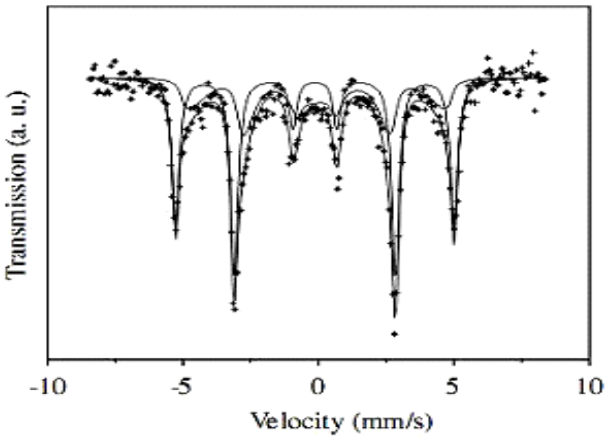

Figure 1: Mössbauer spectra for $[Fe(40)=Cu(20)]_{20}$ Multilayers with $M_{Fe}=2.2\mu B$, recorded at room temperature.

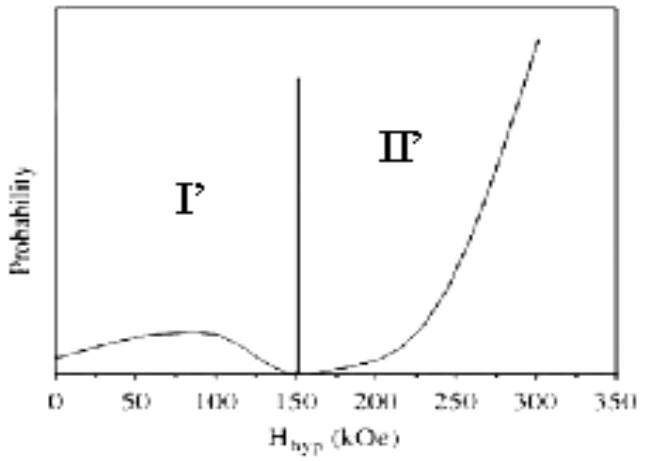

Figure 2: Hyperfine field distribution for $[Fe(40)=Cu(20)]_{20}$, recorded at room temperature, relative to the broad component.

This component represents *33%* of the total area of the spectrum, and reveals two parts labelled (*I'*) and (*II'*). Part (*I'*) has a form similar to the distribution of *Fe* atoms with low magnetic moments near the *Cu* layer, and part (*II'*) related to the domain starting near the standard *bcc* iron hyperfine field down to *150kOe* which the linear decrease of the hyperfine field may be related to a variation of the conduction electron polarization term when going inside the core iron from the *Fe*/*Cu* contact zone. The relative weights of parts (*I'*) of the distributions allowed estimating the equivalent thickness of the iron layer representing atoms with low magnetic moments.

III. Model and simulation technique

We consider for reason of simplicity a simple cubic multilayered system made up of n Magnetic and Non-Magnetic atomic planes which are $L\times L$ in cross section. The classical Heisenberg Hamiltonian of the system is:

$$H = -\sum_{<i,j>}^{N} J_{ij}\left(S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z\right),$$

Where N is the number of sites, S_i^x, S_i^y, S_i^z are spin operators following the axes of space *OX*, *OY* and *OZ* respectively at site i and sum is taken over nearest-neighbours pair of spins. J_{ij} denotes nearest-neighbour exchange interaction between S_i and S_j spins.

Where $\vec{S}_i$ is the classical Heisenberg spin, J_{ij} is the nearest-neighbour exchange interaction sites i and j. We define the magnetization as follows:

$$M(T) = \left\langle \left[\left(\sum_i m_i^x\right)^2 + \left(\sum_i m_i^y\right)^2 + \left(\sum_i m_i^z\right)^2 \right]^{1/2} \right\rangle_T / N,$$

Where the N ($N=L\times L\times n$) is the number of atoms, $m_i^x = -g_i S_i^x/(g_i - 1)$ is the x component of the moment of the atom i in Bohr magneton units, and $\langle \rangle_T$, which means statistical average at temperature T.

The flowing magnetic parameters are those of free atoms Magnetic: $gM=gFe=2$, $S_M=S_{Fe}=1$ and $SNM=SCu=0$. The exchange interactions have been adjusted to obtain pure polycrystalline *Fe* Curie temperature (see ref [5]).

We describe realistic system with real interface in the sense that is not really sharp but exhibit a certain degree of defects and interdiffusion. Although *Fe* and *Cu* are insoluble materials, only ~ *4% Fe* dissolves into *Cu* and ~ *10% Cu* into *Fe* [6], *Fe* and *Cu* can nevertheless form a mixed interface layer under certain conditions of synthesis. There are also some reports on this system showing that alloying effect may be obtained between *Fe* and *Cu* layers during preparation with different techniques [7-10]. We consider as model geometry, a multilayer system consisting of *M* and *NM* layers and a disordered interface. The interface atomic model is characterized by some arrangement of *M* and *NM* atoms so that ($M_{1-x}NM_x$) is a three-dimensional alloy with $0\le x \le 1$ ($x=0$ or $x=1$ abrupt interface, $x\neq 0$ interface alloys). We have limited the interdiffusion to two atomic layers (this is due to the weak miscibility between *Fe* and *Cu*). We consider the interface made up of two areas; an atomic layer beside the *Magnetic* layer rich in *Magnetic* element and poor in *Non-Magnetic* element ($M_{k'}NM_{1-k'}$) with $0.5\le k' \le 1$ and the other layer adjacent to the *Non-Magnetic* layer rich in *Non-Magnetic* element and poor in *Magnetic* element (M_kNM_{1-k}) with $0\le k\le 0.5$ (Figure 3). k (or k') is related to the abundance of element *Fe* in each of the two areas for a fixed composition x [5,11]. Such interface configurations are predicted in real samples (*Fe/Cu*) analyzed experimentally, using Mössbauer spectroscopy [4, 12-16].

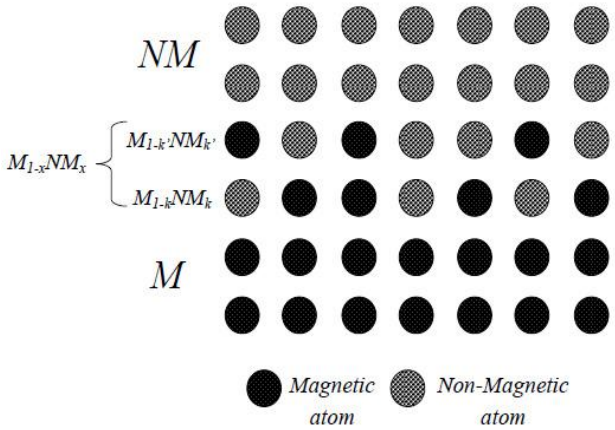

Figure 3: Schematic representation *2D*-dimensional cross-section through the *M/NM* multilayers system

The numerical method is the importance sampling MC procedure at each temperature based on the standard Metropolis algorithm [17] based on random trial step [18]. The thermodynamic equilibrium at each temperature is corresponding to the minimization of the free system

energy of the (Figure 4). In this algorithm, the spins are examined individually. A site *i* is randomly chosen and a unit vector defined by the random choice with uniform distribution of its *z*-component $z_i \in [-1, 1]$ and its azimuth angle $\varphi_i \in [0, 2\pi[$ is determined.

It has to be noted that this spin trial rotation procedure is isotropic. Then the energy variation ΔE associated to this rotation is calculated. The next step is the following:

– if $\Delta E \leq 0$, the rotation is accepted;

– if $\Delta E > 0$, the rotation may be accepted with a probability that is proportional to the Boltzmann factor $exp(\Delta E / k_{BT})$ in order to take into account thermal fluctuations.

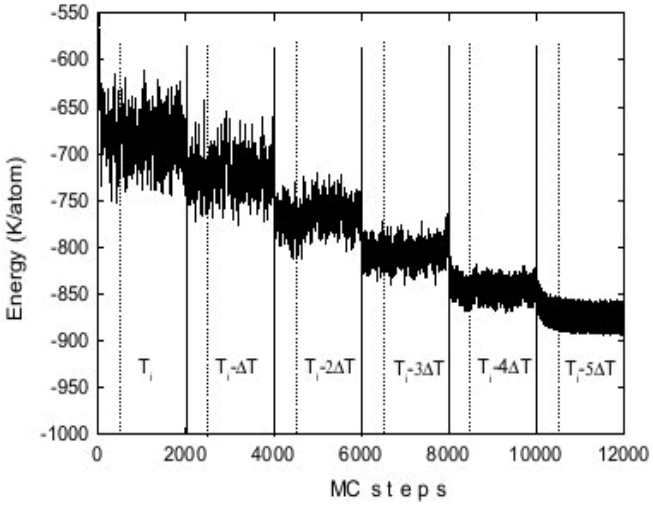

Figure 4: Diagrammatic representation of the variation of energy during the process of simulated annealing, the temperature decrease of ΔT.

One MC step consists in examining all spins of the system once. At each temperature, 5×10^3-7×10^3 MC steps were performed to reach thermodynamic equilibrium, and afterwards physical quantities were measured by averaging over the next 1.5×10^4-2.5×10^4 MC steps.

We calculate magnetization distribution from our MC simulations in the following way: at a temperature comparable to that of the Mössbauer spectroscopy measurement, we waited until the system has attained thermal equilibrium, which has been controlled by usual order parameters. In thermal equilibrium the determination of fluctuation spin rate denotes, at a given time window, computing the average magnetization value <M> at every spin interchange. This can be done efficiently by storing the time at which spin was flipped and then updating average when the next flip takes place. The time window is related to the number of MC steps, i.e.; number of attempt interchanges per spin. In agreement with G. Bayreuther [19], the macroscopic magnetization is proportional to the measured hyperfine field. Because the copper layer is nonmagnetic, the equivalence macroscopic magnetization-hyperfine field is very realistic. So, the hyperfine field distribution can be deduced. Then we proceed by a Gaussian fit in the same way as for experimental data.

It is important to note that finite size effects have no significant influence on the physical properties under consideration. Only nearest-neighbour interactions should be taken into account since magnetic studies of such systems in particular in amorphous alloys, shows evidence that the value of the next nearest-neighbour interactions is one order of magnitude smaller [20]. Indeed, the exchange interactions are very strong in transition metal but at short range.

IV. Numerical results

In our simulations, we have a bilayer made up of eight *Magnetic* planes, three *Non-Magnetic* planes (M_8/NM_3) ($n=n_M+n_{NM}=11$) and we take the interface in the form of homogeneous amorphous ($M_{0.5}NM_{0.5}$) with $k+k'=1$. Several lateral dimension sizes are considered ranging from 10×10 to 30×30 atomic sites. The interface spin exchange interaction ($J(i)$) is set equal to the bulk spin exchange interaction ($J(b)$). The adapted sizes matches the *Fe(40Å)/Cu(20Å)* bilayers considered in the experiment subject of the comparison. We note that the interface is composed of different types of magnetic atoms that it is possible to discern by their nearest neighbour (nn) numbers of magnetic atoms. In a case with *20x20* spins at each atomic monolayer layer, there are 0.*2%* magnetic atoms with *0* nn, *1.4%* with 1 nn, *3.2%* with *2* nn, 3.8% with 3 nn, 3.7% with 4 nn, 8.7% with 5 nn and 2.8% with 6 nn magnetic atoms. In sum there are *23.8%* magnetic atoms at the interface from the total of magnetic atoms in the bilayer. At the magnetic layer, all the atoms are surrounded by magnetic atoms (*6* nn). Furthermore, the arrangement into clusters correlated to the nn is an important criterion in analysing experimental result.

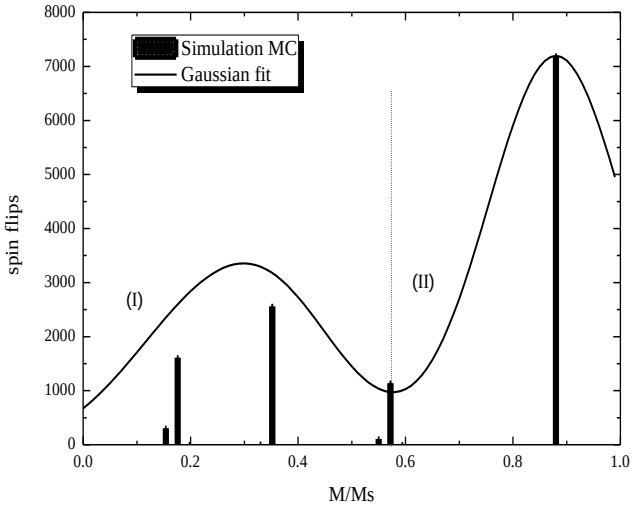

Figure 5: The number of spin flips in the whole system (M_8/NM_3) during 2.1×10^4 MC steps and corresponding Gaussian curve fit at room temperature in function of magnetization distribution, with $k=0.30$ ($k'=0.70$)

To examine the fluctuation behaviour of all the spins in the sample over a suitable time interval as a function of interface morphology and temperature. The number of flips during 2.1×10^4 MCS is plotted in figure 5. It is possible to discern distinct groups of spins depending on the magnetization of the layer where are located. Since interaction energy for a given spin is proportional to the number of nearest neighbours, the small number of spins with fewer magnetic nearest neighbours flips on average *1000-2500* times at the interface. In the magnetic layer with a larger number of spins, we can note until *7500* flips. The peak positions are attributed to the magnetisation of each layer; i.e. the interface *(part I)* or the magnetic layer *(part II)*. In figure 5, we have drawn a corresponding Gaussian curve fit to the spin flips distributions. The Gaussian profile is performed in order to: i) reflect approximately the broad atomic distribution at the interface around the composition *x* when a multilayer is considered rather than a bilayer, ii) take into account the distribution of the interatomic distances in real samples which influence the exchange interactions.

Figure 5 shows that we have relatively succeeded in reproducing experimental curve of hyperfine field distribution shown in figure 2. The variation are attributed to two parts of distribution tagged *(I)* and *(II)* [4]. The part *(I)* related to the domain *[0-0.6]* corresponds to *magnetic* atoms with low magnetization involved in the interface and part *(II)* related to domain *[0.6-1.0]*. In this last range, the variation of the magnetization is attributed to the *magnetic* atoms located in the centre of iron layer who do not feel the effect of the interface.

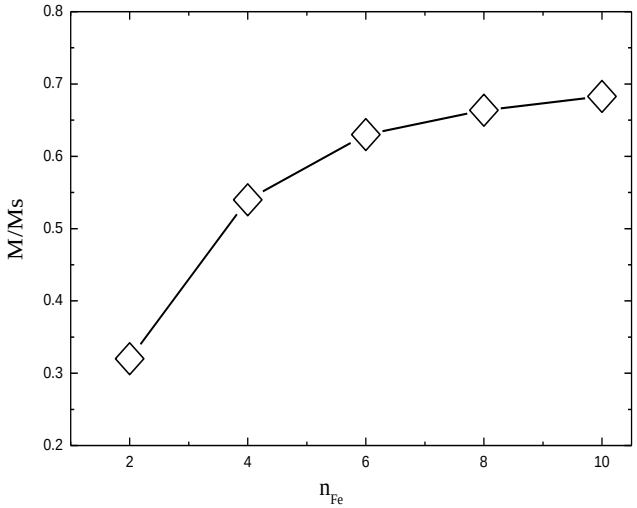

Figure 6. Evolution of the reduced magnetization (M/M_s) versus the number of iron layers n_M. M_s denotes the saturation magnetization.

In what follows, we investigated the effect of *magnetic* thickness on the magnetization. Figure 6 shows how the fluctuation behaviour affect the average magnetization at room temperature for n_M=*2, 4, 6, 8* and *10* planes. It is clear that the magnetization (M/M_s), versus the number of plane of magnetic layer n_M, remains almost constant for *6-10* planes and decreases for n_M <6.

In a logical sequence, we note that the composition of the interface alloys ($M_{1-x}NM_x$) has an effect on the variation of magnetization for the whole system $(M/NM)_{11planes}$. Figure 7 exhibits the variation of spins flipped at room temperature in function of reduced magnetization for a series of interface composition; *x=0.25, 0.50* and *0.75*. Increasing the non-magnetic atoms rate at the interface reduces the number of spins at the interface, and consequently the interface magnetization. Therefore, the spins flipping distribution is shifted to the left. The second peak position relative to the magnetic layers remains at the same position and the number of flipped spins is barely affected.

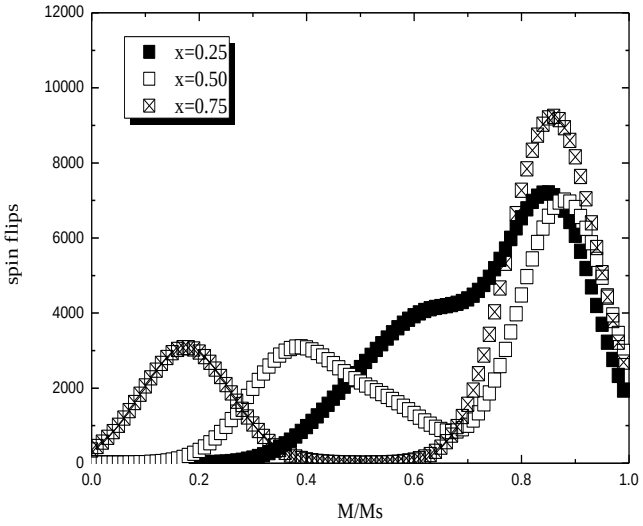

Figure 7: Variation of spin flips for the whole system M_8/NM_3 at room temperature as function of magnetization distribution for concentrations (*x=0.25, 0.50* and *0.75*) of the interface alloys ($M_{1-x}NM_x$).

V. Conclusion

In this work, Monte Carlo simulation and multilayer Heisenberg models have been used to investigate the hyperfine field distribution in Magnetic/Non-Magnetic (*M/NM)* multilayers, we were able to reproduce the same experimental results on *Fe/Cu* multilayers and to develop a fundamental understanding of the structure and morphology dependence of the magnetic properties. Our simulations confirm that layer thickness of magnetic, concentration of interface alloys ($M_{1-x}NM_x$) modifies the magnetic properties of the *M/NM* multilayers. The results are qualitatively in good agreement with those observed experimentally. Finally, it should be noted that all the results we find for the system Fe/Cu are similar to other systems such as Co and Ni.

VI. References

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