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Onset of magnetic order in multilayers of Fe and Ni on and embedded in fcc-Cu(100) substrates

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ABSTRACT

We investigate magnetic correlations and local magnetic moments at finite temperatures in some Fe and Ni multilayers on and embedded in Cu(100). Our study is based on a mean-field theory of magnetic fluctuations for layered materials within the first-principles local spin-density functional theory. We find that although there is no significant local moments formation in the Ni layers, the Ni layers are not magnetically dead in the sense that they mediate magnetic interactions between Fe layers separated by Ni layers. The Curie temperature of Fe_nNi_m/Cu(100) and Ni_nFe_m/Cu(100) films is almost independent of the Ni layer thickness, however, in $Fe_1Ni_nFe_1$/Cu(100) and $Fe_1Cu_nFe_1$/Cu(100) films, the Curie temperature exhibits an interesting oscillatory behaviour as the spacer layer thickness is increased. We show that there is a connection between this behaviour and the spin-reorientation transition observed in some Ni films. In the $Ni_1Fe_nNi_1$/Cu(100) and Cu_1Fe_n/Cu(100) films we found that the nature and strength of the magnetic correlations depends upon the Fe layer thickness.

1. Introduction

Among the transition metal ferromagnetic systems, Ni and materials containing Ni are the most intriguing. Compared to other ferromagnetic transition metals, fcc-Ni exhibits a very small magnetic moment below the Curie temperature: bcc-Fe has a magnetic moment of $2.2\mu_B$ and a Curie temperature of 1043 K [1], hcp-Co has a magnetic moment of $1.58\mu_B$, fcc-Co has a magnetic moment of $1.61\mu_B$ and a Curie temperature of 1388 K [2], but fcc-Ni has a magnetic moment of $0.62\mu_B$ and it becomes ferromagnetic only below 631 K [1]. Furthermore, while bcc-Fe is a weak ferromagnet, fcc-Ni is a rather strong ferromagnet. Another major difference is that, Fe and Co films on Cu(100) exhibit larger magnetic moments compared to their respective bulk values, whereas the magnetic moment of fcc-Ni/Cu(100) films is substantially smaller compared to its value in the bulk [3–5]. No magnetic domains are found in films of thickness less than 6 monolayers [6] which indicates that the films are paramagnetic. Thicker films are ferromagnetic but still exhibit quite small magnetic moments [7,8]. However, some reports claim that the magnetic moments in Ni/Cu(100) films are comparable to that of the bulk Ni [9]. But, above the Curie temperature, because of magnetic moment fluctuations, the local magnetic moments in Ni/Cu(100) films are expected to be very small.

The rich magnetic properties of Fe/Cu(100) and Ni/Cu(100) films make it quite tempting to create novel magnetic films of Fe and Ni on Cu(100) substrates with 'tailored' magnetic properties. There has been great interest recently in Fe and Ni based ferromag netic-nonmagnetic-ferromagnetic trilayer systems because of their potential implications in weak magnetic field detectors and related technologies, such as for use as magnetic biosensors. The FeNi/Cu/FeNi trilayers have been at the forefront of this technology at the moment because they offer good magnetoimpedence effect over a wide range of frequencies [10–12]. Studies on multilayers of Fe, Ni and Cu with similar geometry as the aforementioned trilayers can help understand the mechanism of magnetic properties in these films and hence can facilitate the design of such technologically important devices. Also interesting is the exhibition of spin-reorientation transition (SRT) in the Fe and Ni multilayers on Cu which depends upon the thicknesses of Ni and Fe layers. The Ni-layer thickness at which the SRT takes place increases in the presence of an Fe underlayer [13]; in the Ni_nFe_6/Cu(100) films, the in-plane to out-of-plane transition takes place for n between 12 and 15 monolayers [7] compared to the thickness between 7 and 10 for Ni_n/Cu(100) films. This is quite peculiar because Fe_6/Cu(100) films are known to exhibit perpendicular magnetic anisotropy, which should help to build up and stabilize an out-of-plane magnetisation for the Ni overlayer. It is believed that the antiferromagnetic nature of the Fe underlayer is responsible for the increase in the thickness of Ni layer for the SRT [6]. It is reported [6] that for

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Fe layer thickness between 5 and 11 monolayers in the Ni_nFe_m/Cu (100) films the Fe layer is antiferromagnetic with a ferromagnetic live layer at the Ni interface. However, if Fe layers are deposited on the Ni layers as capping, perpendicular anisotropy is established for thinner Ni films compared to a bare Ni film as expected [14].

In a previous study [15], we had presented the local magnetic moments on different layers of Ni_n/Cu(100), Ni_nFe_m/Cu(100), Fe_nNi_m/Cu(100) and $Fe_1Ni_nFe_1$/Cu(100) films as well as their Curie temperatures. We had found no significant local moment formation in the Ni layers in any of the films in confirmation of the experimental observations [16–18]. However, these Ni layers were not magnetically dead layers because they were seen to mediate magnetic correlations between the Fe layers close to the Ni layers, thus confirming the findings of Wilhelm et al. [18]. The Curie temperatures of the films were found to be influenced quite strongly by the presence and layout of Ni layers in the film. In particular, the Curie temperature of $Fe_1Ni_nFe_1$/Cu(100) film was found to oscillate when the Ni layer thickness was increased from one monolayer to seven monolayers. In this work, we present a more detailed study of the magnetic properties of Ni_nFe_m/Cu(100), Fe_nNi_m/Cu(100) and $Fe_1Ni_nFe_1$/Cu(100) films. We also present studies on some new films, such as $Fe_1Cu_nFe_1$/Cu(100), $Ni_1Fe_nNi_1$/Cu(100), and Cu_1/Fe_n/Cu(100) films as well as the case of one and two monolayers of Ni burried in fcc-Fe on Cu(100) substrate. We have calculated the intralayer and interlayer magnetic correlations in each film from which we can determine the paramagnetic spin susceptibility at different temperatures and obtain the Curie temperatures.

We have used a *first-principles* electronic structure method based on the spin-density functional theory to study the magnetic properties of multilayers in their paramagnetic phase above the Curie temperature. Over the last decade, much progress has been made in the *first-principles* theories of itinerant electron magnetism at finite temperatures [19,20]. In our study, the spin fluctuations above the Curie temperature are modelled as 'disordered local moments' (DLM) [21]. This model is easily incorporated into a *first-principles* electronic structure scheme [22,23] and has been very successful for bulk solids [24,25]. For layered systems, an *ab initio* electronic structure based theory incorporating the DLM [26–30] is implemented by using the screened Korringa-Kohn-Rostoker (SKKR) coherent-potential approximation (CPA) method [31]. This theory has also been extended to study the temperature dependence of magnetic anisotropy [32–34] by generalising it to take the relativistic effects into account.

The magnetic moments and Curie temperatures of Ni_nFe_m/Cu (100), Fe_nNi_m/Cu(100) and $Fe_1Ni_nFe_1$/Cu(100) films were presented in our previous work [15]. In this work, we present the interlayer and intralayer magnetic correlations between the Fe layers as well as the paramagnetic spin susceptibility of these films. A comparison of the interlayer magnetic correlations of these films with those of the bare Fe_n/Cu(100) films shows that a Ni underlayer or overlayer changes the nature of interlayer magnetic correlations only between the two Fe layers closest to the Ni layer. We also find that the difference in the Ni layer thickness for SRT in the Ni_n/Cu (100) and Ni_nFe_m/Cu(100) films [6–8,13,14] is related to the difference in the nature of the interlayer magnetic correlations because of a Fe underlayer or overlayer. In the $Fe_1Ni_nFe_1$/Cu(100) films, the Curie temperature was found to oscillate when the Ni layer thickness was increased from one monolayer to seven monolayers [15]. In the $Fe_1Cu_nFe_1$/Cu(100) films the Curie temperature is somewhat stable as a function of Cu spacer thickness. We also find that the interlayer magnetic correlation between the Fe layers exhibits an oscillatory spacer-thickness dependence for the Cu spacer while it remains ferromagnetic for Ni spacer until it is 6 monolayers thick. In the $Ni_1Fe_nNi_1$/Cu(100) and Cu_1/Fe_nCu(100) films the nearest-neighbour interlayer magnetic correlations between the Fe layers are antiferromagnetic in nature. The Curie temperatures of the Cu_1Fe_n/Cu(100) films exhibit an oscillatory thickness dependence, but the Curie temperatures of $Ni_1Fe_nNi_1$/Cu(100) films do not show any oscillatory behaviour.

The paper is organized as follows. In Section 2 we briefly outlines the computational details, in Section 3 we present our results and in Section 4 we present the conclusions.

2. Computational details

Our study of finite temperature magnetism of multilayers is based on the DLM model adopted into the *first-principles* SKKR-CPA method. For details the reader is refered to Ref. [27]. The key quantity to these calculations is the layered-resolved paramagnetic spin susceptibility at temperature T, given by [27]

$$\chi_{PQ}(\mathbf{q}_{\parallel}) = \mu_P^2 \left[(3k_BT)I - S^{(2)}(\mathbf{q}_{\parallel})\right]^{-1}_{PQ} \tag{1}$$

where $\mathbf{q}_{\parallel}$ is a wave-vector in a layer, μ_P is the magnitude of the local magnetic moment in layer P, and k_B is the Boltzmann constant. The matrix elements $S^{(2)}_{PQ}(\mathbf{q}_{\parallel})$ are the two-dimensional lattice Fourier transforms of the 'direct correlation functions' [27]. Within a non-relativistic electronic structure framework, the DLM model maps onto a random binary alloy $A_{0.5}B_{0.5}$ with the species A and B representing the "up-spin" and "down-spin" sites respectively. So, thermal spin fluctuations can be handled using the "CPA" for layered systems [31]. With this approximation, $S^{(2)}_{PQ}(\mathbf{q}_{\parallel})$ are expressed as convolution integrals in the surface Brillouin zone. However, to study ferromagnetic transitions, we only need $S^{(2)}_{PQ}(\mathbf{q}_{\parallel})$ for $\mathbf{q}_{\parallel} = 0$, which is related to the change in the "Weiss field" $S^{(1)}_P$ in layer P with respect to a change in the magnetisation on layer Q, [27]

$$S^{(2)}_{PQ}(\mathbf{q}_{\parallel} = 0) = -\frac{\partial S^{(1)}_P}{\partial m_Q} \tag{2}$$

When the system is cooled down from a high temperature, the ferromagnetic order starts around the temperature at which the instabilities in the spin fluctuations diverge. So at the Curie temperature,

$$\|(3k_BT_c)I - S^{(2)}(\mathbf{q}_{\parallel} = 0)\| = 0 \tag{3}$$

This means that the largest positive eigenvalue of $S^{(2)}(\mathbf{q}_{\parallel} = 0)$is related to T_c as,

$$3k_BT_c = \text{Largest positive eigenvalue of } S^{(2)}(\mathbf{q}_{\parallel} = 0)$$

In our calculations, all the films are treated as perfect fcc structures (lattice parameter, 6.83 a.u.) deposited layer by layer on the substrate ignoring interdiffusion and lattice strain. For each film, the electronic structure of each layer of the film, as well as that of a buffer of three layers of the substrate and a buffer of three layers of vacuum were calculated self-consistently. The local charge densities were calculated within the atomic-sphere approximation in which all the atoms were assumed to have the same atomic radii. The Brillouin zone integration was performed by using 45 $k_{\parallel}$ points in the irreducible part of the surface Brillouin zone.

3. Results and discussion

In this section we discuss our results. In subsection 3.1 we present the results for Ni_nFe_m/Cu(100) and Fe_nNi_m/Cu(100) films, and in subSection 3.2 we present the results for $Fe_1Ni_nFe_1$/Cu(100) and $Fe_1Cu_nFe_1$/Cu(100) films. In subSection 3.3 we present the results for $Ni_1Fe_nNi_1$/Cu(100) and Cu_1Fe_n/Cu(100) films and in subSection 3.4 we present the results for Ni layers burried in Fe/Cu(100).