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Noncollinear magnetic structures of FeMn ultrathin films on Cu(001)

B.R. Malonda-Boungou [a, b], N. Binggeli [b, *], B. M'Passi-Mabiala [a]

[a] Groupe de Simulations Numériques en Magnétisme et Catalyse, Faculté des Sciences et Techniques, Université Marien Ngouabi, PB 69 Brazzaville, Congo
[b] The Abdus Salam International Centre for Theoretical Physics, Strada Costiera 11, 34151 Trieste, Italy

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ABSTRACT

We investigate, by means of *ab initio* spinor-density-functional calculations, the magnetic structures of one and two monolayers of MnFe ordered alloys on the Cu(001) surface. For comparison, we also examine the FeMn compound with $L1_0$ structure, as bulk reference limit of the FeMn films. In the limit of the ultrathin films considered, the 3Q antiferromagnetic ground state of the FeMn bulk is unstable. A strongly non-collinear planar ferro- or ferri-magnetic spin structure is established, instead, as ground state. It is characterised by mainly parallel alignments of the Fe spins and Mn-Fe spins with orientations close to perpendicular. Depending on the Mn/Fe chemical order and coordination, the Fe-Mn exchange coupling is either antiferromagnetic or ferromagnetic biased. The trends of the spin structure and Fe-Mn exchange coupling with different chemical orders and thicknesses of the films are discussed based on the calculated Fe and Mn local density of states. In particular, the behaviour of the Fe-Mn exchange coupling is found to be controlled mainly by the position of the Fermi energy within the Fe-3*d* minority-spin local density of states.

1. Introduction

The fcc-$Fe_{0.5}$ $Mn_{0.5}$ alloy (γ-FeMn) is a noncollinear antiferromagnet with a high Néel temperature (T_N ~ 500 K), used as exchange bias material in magnetoresistive devices [1,2]. The magnetism of γ-FeMn has been the subject of numerous investigations over more than 5 decades. In spite of its importance, however, the precise non-collinear antiferromagnetic (AFM) structure of bulk γ-FeMn has remained fairly controversial until recently [3].

In the past 1-2 decades, much research attention has been focused on the magnetic properties of γ-FeMn in the form of ultrathin films grown either directly on Cu(001) or on a Co or Ni ferromagnetic (FM) layer deposited on Cu(001) [4–15]. There is a very good lattice matching between FeMn and Cu (~0.4% misfit) [16], which makes Cu an ideal substrate for epitaxial growth of MnFe layers and related FeMn-based single crystalline heterostructures. Such single-crystalline heterostructures, with well-defined interface structural properties, are particularly convenient model systems to study and engineer at the atomic scale the magnetic coupling across the FM/FeMn interface [17–22]. In this connection, the knowledge of the magnetic structure of ultrathin FeMn films on Cu is an important prerequisite.

* Corresponding author.
E-mail addresses: malonda_brice@yahoo.fr (B.R. Malonda-Boungou), binggeli@ictp.it (N. Binggeli), bmpassimabiala@gmail.com (B. M'Passi-Mabiala).

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In the bulk, chemically disordered γ-FeMn adopts a three-dimensional (3D) noncollinear AFM structure, the so-called 3Q structure [3]. When grown as an epitaxial film, however, at sufficiently low thickness a different ground-state spin structure may occur due to the reduced dimensionality. This might be expected, e.g., considering the various strongly non-collinear magnetic structures discovered in other low-dimensional/nanostructured Mn-Fe systems [23–27]. Such non-collinear spin structures have been disclosed by recent first-principles computational studies for Mn nanostructures on Fe surfaces [23,24], as well as by *ab initio* calculations and x-ray magnetic dichroism experiments for a Mn monolayer (ML) on the Fe surface [25,26], and by single-crystal neutron diffraction for Mn/Fe nanostructured multilayers [27]. *Ab initio* theoretical studies of such systems have revealed, in particular, Fe-Mn and Mn-Mn couplings which strongly depend on the local environment and geometry [23,25,28,29] and can lead to complex non-collinear magnetic structures often due to magnetic frustrations in low dimensions and nanostructures [23,25,26].

Experimentally, for the FeMn films, temperature-depend magneto-optic Kerr-effect studies have indicated the existence of AFM order in FeMn films with thicknesses above 8 ML [21]. Furthermore, room-temperature magnetic circular dichroism photoelectron emission microscopy and x-ray magnetic linear dichroism have provided evidence of a 3D noncollinear AFM structure (bulk-type) in 15-ML-thick FeMn films [7] as well as indications of a possible AFM structure for FeMn films with thicknesses above 7 MLs [7,18,30]. The Néel temperature of FeMn films, however, decreases with decreasing thickness, reaching room temperature at about 9 ML [6,21]. The ground-state spin structure (at low temperature) of FeMn films with thickness below 7 MLs has not been established experimentally.

For Co/FeMn bilayers on Cu(001), X-ray magnetic circular dichroism studies have shown that the Fe magnetic moments at the Co/FeMn interface align ferromagnetically with respect to the Co moments [8–10]. By means of non-collinear-spin density functional theory (DFT) calculations, Nakamura et al. [11] investigated the magnetic structure of a $(Co)_3/(FeMn)_7(001)$ superlattice. Their study indicated that the Fe moments at the Co/FeMn interface reorient away from their directions in the bulk FeMn so as to be parallel to the Co-moment direction, consistent with the experimental observation [8–10]. For the $(Co)_3/(FeMn)_7$ superlattice, this local spin reorientation at the interfaces resulted in a two-dimensional (2D) noncollinear arrangement of the spins in the FeMn film, with Fe spins pointing along the Co magnetisation direction and canted Mn moments with magnetisation antiparallel to the Fe and Co magnetisations [11]. The occurrence of this 2D spin configuration in the FeMn layer was ascribed to the effect of the interaction with the FM Co layer in the *ab initio* study [11]. We will show here, based on similar *ab initio* calculations, that a strongly noncollinear 2D spin configuration is also established as ground state with reduced thickness of the MnFe epilayer, in the absence of a FM layer.

Theoretically, DFT-based studies of the magnetic properties of ultrathin FeMn epilayers on Cu or on a non-magnetic substrate (in the absence of a FM layer) have been limited so far to collinear-spin investigations [14,15,31]. This includes a study of the chemically ordered $c(2 \times 2)$ FeMn ML on Cu(001), which showed the existence of a frustrated AFM structure [14], as well as studies of FeMn epilayers of 1–3 MLs on Cu(001), which indicated FM or AFM Fe-Mn nearest-neighbour coupling, depending on the local coordination and geometry [15]. A study of 1 ML of FeMn on the bcc-W(001) surface has also been performed (corresponding to a 24% expansion of the in-plane lattice constant of the FeML ML with respect to the Cu(001) surface), which displayed a competition between an AFM and a frustrated ferrimagnetic ground state [31].

In the present study, we address by mean of first-principles non-collinear-spin DFT calculations, the ground-state magnetic structure of FeMn epilayers on Cu(001) in the limit of ultrathin, one-to two- ML-thick, FeMn films. We also examine, for comparison, the lowest-energy spin configurations of the bulk FeMn compound with $L1_0$ atomic order (i.e., an fcc(001) $(Fe)_1/(Mn)_1$ superlattice), as bulk limit of the chemically ordered FeMn films. We consider different types of chemical ordered FeMn MLs and a Mn(1-ML)/Fe(1-ML) bilayer on Cu(001). In all cases, a similar type of 2D strongly noncollinear Fe-Mn spin configuration is found as ground state of the 1–2 ML FeMn films, with either slightly ferro or antiferro magnetic alignment between the Fe and Mn magnetisations, depending on the Fe/Mn chemical order and coordination. The behaviour of the spin structure and trends of the Fe-Mn exchange coupling, when going from the FeMn $L1_0$ bulk, to the bilayer, and to the ML, are rationalised based on the analysis of the corresponding Fe and Mn spin-resolved local density of states.

The manuscript is organised as follows. The next section is devoted to the computational details. The calculated lowest-energy magnetic states of the FeMn $L1_0$ bulk, 1-ML Mn/1-ML Fe/Cu(001), and 1-ML-FeMn/Cu(001) systems are presented in Sections 3.1, 3.2, and 3.3, respectively. The trends of the non-collinear magnetic structures and Fe-Mn exchange couplings, from the bulk, to the bilayer, and to the ML, and with different chemical orders, are discussed in Section 3.4, followed by the conclusions in Section 4.

2. Computational model

The calculations were performed within density functional theory (DFT). We used the pseudopotential plane-wave method, as implemented in the Plane Wave self-consistent field (PWscf) code distributed with the Quantum ESPRESSO package [32]. The unconstrained spinor DFT was used to deal with noncollinear spin structures. Unless otherwise specified, the magnetic structures were obtained within the local spin density approximation (LSDA), using the exchange-correlation functional parametrized by Perdew and Zunger [33]. In a few cases, for collinear spin structures, we also carried out computations within the generalized gradient approximation (GGA), using the Perdew-Burke-Ernzerhof functional [34]. All calculations were performed with scalar-relativistic Vanderbilt ultrasoft pseudopotentials [35], using a plane-waves kinetic energy cutoff of 30 Ry. The 3*d* and 4*s* states of copper, iron, and manganese were treated as valence states. The non-linear-core correction to the exchange-correlation potential was included for all three elements.