Fe epilayer magnetic features induced by a covalent, magnetic and metallic substrate

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Abstract.

In recent years there has been theoretical and experimental interest in the properties of Fe thin layers grown on metallic substrates. We present here a theoretical study based on \textit{ab initio} calculations of the structure and magnetic characteristics for a very thin Fe film grown on an As-terminated MnAs substrate, which is metallic. Particular emphasis is put into the relaxation and reconstruction of the interface and on the characterization of the magnetic coupling with the substrate. The absence of Fe interdiffusion and the presence of an intermediate As layer ensure the partial magnetic decoupling between epilayer and substrate.

1. Introduction

Thin films of different kinds of materials with potential electronic, magnetic and spintronic applications, along with other functionalities, have been investigated during the last decades because of their importance and interest in the development of micro- and nanotechnological devices. Among them, manganese arsenide (MnAs) thin films have been studied quite intensively in the last years due to the good compatibility of this material with standard semiconducting substrates\cite{1, 2, 3} and also due to its high spin polarisation\cite{3}, both characteristics being well suited for the development of spintronic devices\cite{1, 4}.

MnAs is, in fact, a metallic material which presents an interesting variety of polymorphic transformations as a function of temperature\cite{1, 5}. In bulk, the low temperature hexagonal structure of the NiAs-type (\(\alpha\) phase) is ferromagnetic and it has a first order transition at 40\(\degree\)C, where the system evolves into a paramagnetic orthorhombic structure of the MnP-type (\(\beta\)-phase). At 127\(\degree\)C, it undergoes a second order transition transforming again into a low temperature hexagonal structure, which is paramagnetic, namely the \(\gamma\) phase. Most of the latest works on MnAs have been done for thin films of this material grown on GaAs(001).

In a temperature window around room temperature, whose spread depends on the MnAs film thickness, the contact between GaAs and MnAs gives rise to a strain-driven pattern of coexisting and alternating magnetic and non-magnetic MnAs stripes\cite{1, 2, 4, 5}. More recently, Fe epilayers of 5 to 10 nm were grown on top of MnAs(1100)/GaAs(001) in order to tune the period of the above mentioned stripes\cite{6}, or to use these heterostructures as magnetic active templates as a function of temperature\cite{7}. In Ref. \cite{7} it has been
reported the magnetic response of these Fe epilayers when the MnAs thin films go from the α to the β phase, through the phase coexistence temperature region. A magnetic decoupled behaviour of the Fe epilayers was observed, characterized by the fact that the magnetization could be either parallel or anti-parallel to the MnAs substrate. This behaviour, revealed a weak magnetic coupling between both magnetic materials. This result was supported by FMR studies, where the evolution of the magnetic anisotropies of the combined system Fe(5nm)/MnAs(100nm)/GaAs(1mm) as a function of temperature was measured. It was also found that, outside the MnAs coexistence temperature range, the Fe films are also magnetically decoupled from the substrate.

The observed magnetic behaviour of the Fe epilayers should be traced back to interfacial effects, but the characteristics and nature of the structure at the interface of an ultrathin Fe epilayer have not yet been approached. Actually, this interface was only characterized for Fe overlayers of thickness beyond 30nm, for which a mixed MnFeAs antiferromagnetic compound in the interfacial region was found. The antiferromagnetic structure of this interfacial compound made it possible to explain the magnetic decoupling between the Fe films and the MnAs substrates when the thickness of the overlayer competes with the thickness of the MnAs substrate. For thinner Fe overlayers, less than 10nm, this discussion is still open and, in this work, we try to give a partial answer to it. We theoretically study the structure of a Fe film of approximately 4Å grown on an As-terminated MnAs substrate. Particular emphasis is put into the relaxation and reconstruction of the interface, which characterize this ultrathin film regime.

2. Results and discussion

Electronic calculations are performed within the framework of density functional theory as implemented in the VASP code. PAW pseudopotentials are used with the choice of exchange and correlation potential within the Generalized Gradient Approximation (GGA) as parametrized by Perdew et al. We consider a plane waves basis set with kinetic energy values of up to 350 eV, and a 20×20×1 grid in k-space. The internal coordinates are fully relaxed until forces are smaller than 0.04 eV/Å.

Regarding the crystal structure of the MnAs substrate, in our calculations we consider the experimental data which states that α-MnAs is hexagonal (B3\(_1\)) and that it grows epitaxially along the (1100) direction. We simulate the ferromagnetic α-MnAs substrate using three

Figure 1. Three Fe layers on MnAs(1100). a) Schematic lateral view of the relaxed structure. b) Charge isosurface plot corresponding to 0.05 e/Å\(^3\), which emphasizes the Fe-As bonds.
Figure 2. Projected densities of states (PDOS) on a) Fe, b) As and c) Mn atoms, at the interface of the 3L-Fe/MnAs heterostructure (with red lines) and the PDOS projected on the same atoms for the corresponding bulk (shown with shadow areas). The energy values are referred to the Fermi level.

MnAs unit cells separated by a vacuum region of 10 Å and perform periodic slab calculations with in-plane cell parameters $a = 3.73$ Å and $b = 5.69$ Å$^3$. No inversion symmetry is imposed in order to have full formula units of MnAs in the unit cell of the slab, and as a consequence, the two free surfaces have different atomic terminations. Therefore, two possible interfaces between Fe and MnAs can be analysed depending on which of the free surfaces the Fe-film is deposited.

We find that the Mn-terminated slab is energetically preferred to the As-terminated one when a Fe overlayer is deposited on them. However, MnAs is usually grown in an As-atmosphere, which favours an As-terminated surface$^2$. We focus, then, in this work on an ultrathin Fe film deposited on the As-terminated substrate.

As indicated by RHEED and TEM characterizations, Fe grows on MnAs(1100) along the Fe(2$\bar{1}$1) direction, perpendicular to MnAs [0001] and [01$\bar{1}$0]$^5$. Following this, our ultrathin film consists of three Fe layers (3L) grown on MnAs along (2$\bar{1}$1), as shown in Fig. 1a). We assume that the interfacial Fe atoms follow the MnAs substrate occupying Mn sites, as it is expected for epitaxial growth conditions. The other two Fe layers keep a bcc-like structure, which is under strain due to epitaxy with the MnAs substrate. The epilayer, as it is usual in epitaxial thin film growth, adopts the parameters of the substrate giving rise to biaxial strain. This clamping propagates throughout the overlayer, it is tensile in one in-plane direction and compressive in the other one. This has already been analysed in Ref.$^{13}$, where it was shown that the biaxial strain contributes to a change in the magnetic anisotropy of the Fe epilayer.

After relaxation of the internal coordinates of the Fe epilayer, the resulting interface is sharp with no Fe interdiffusion, as it is shown in Fig. 1a). The interplane distances among Fe layers are reduced by approximately 20% with respect to the bulk values.

At the interface, the Fe atoms present a magnetic moment similar to the Fe bulk one ($\sim 2.26\mu_B$), while the magnetic moments and charge of the Mn atoms of the MnAs slab do not change in the presence of the Fe epilayer. However, the interfacial As atoms bind covalently with the first and second Fe overlayers. In Fig. 1b) we show a charge isosurface plot of the system and the covalent As-Fe bonds can be clearly recognized.

Regarding the magnetic properties, the lowest energy configuration of the heterostructure under study is ferromagnetic. The difference in energy between the magnetic configuration with the Fe epilayer antiferromagnetically and ferromagnetically aligned with respect to the MnAs substrate is around 10 meV/Fe atom. This value is smaller than the Fe-Fe coupling reported for Fe bulk$^{14}$ and also smaller than the one among Mn atoms in the corresponding MnAs bulk$^3$.

In Fig. 2 we show the projected densities of states (PDOS) of interfacial Fe, As and Mn atoms and also the PDOS for the same type of atoms in $\alpha$-MnAs and Fe bulk structures. We note first, that due to strain the PDOS of Fe in the epilayer presents a smaller average exchange
splitting than the one in bcc Fe bulk, while the PDOS of the interfacial Mn atoms remains unchanged with respect to MnAs bulk. The Mn atoms do not sense, then, the presence of the Fe overlayer, while the interfacial As atoms build covalent bonds with the adjacent Fe atoms, as it can be seen in Fig. 2b) as well as in Fig 1b).

3. Final remarks
We have obtained that the energy difference between ferromagnetic and antiferromagnetic alignments of an ultrathin Fe film grown on As-terminated MnAs is roughly 10 meV, that is equivalent to ~ 110K. This temperature is nearly 200K lower than the temperature of MnAs at phase coexistence, where a magnetic decoupled behaviour was experimentally observed. From our results, this decoupling may be attributed to the absence of Fe interdiffusion and to the presence of an intermediate As layer. The existence of an antiferromagnetic spacer is not needed to achieve the partial decoupling. This goes along with previous results which showed that the magnetic anisotropy energy of this system depends mainly on the substrate induced strain and not on its magnetic state[13].

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