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Magnetocrystalline anisotropy oscillations predicted in Fe/Au(001) superlattices

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Abstract. The magnetocrystalline anisotropy energy in bcc Fe_{2m-1}/fcc Au_5 (001) superlattices ($m = 1$–12) has been calculated from first principles by using a relativistic electronic band-structure method. The calculated magnetocrystalline anisotropy energy in these Fe/Au superlattices is found to oscillate in both sign and magnitude with the Fe slab thickness with a period of about ten monolayers. The origin of this anisotropy oscillation is discussed.

The magnetic anisotropy energy of a magnetic material is defined as the energy difference between the easy- and hard-magnetization axes or the energy required to rotate the magnetization from one direction to another. It determines the low-temperature magnetization direction of the material with respect to its structure, and also influences other magnetic properties of the material such as the spin dynamics and magnetic domain structures. The magnetic anisotropy energy and magnetic moment are the two most important characteristics of a magnetic material. The magnetic anisotropy energy consists of the magnetocrystalline anisotropy energy (MAE) of electronic origin and the magnetostatic anisotropy energy due to magnetic dipole interaction in the material. In a layered magnetic structure, the magnetic dipole energy always 'prefers' an in-plane magnetization, and the possible perpendicular anisotropy must come from the magnetocrystalline anisotropy in the electronic structure via the spin–orbit coupling. Extensive experimental and theoretical studies have been carried out on the magnetic multilayers with the aim of achieving an understanding of the key factors which determine the magnetocrystalline anisotropy of these systems (see, e.g., [1]). This is partly due to the fact that a magnetic multilayer or thin film with a perpendicular magnetization is useful in technological applications such as high-density magneto-optical recording and memory devices (see, e.g., [2]). It has become clear that the reduced dimensionality and symmetry at the transition metal surface and interface in a multilayer can result in a perpendicular anisotropy which is two to three orders of magnitude larger than in the corresponding crystalline solids [1]. It has been found that the perpendicular anisotropy can also be caused by the lattice mismatch strain in a multilayer system (see, e.g., [3, 4]). More recently, Weber *et al* [5] discovered an oscillatory magnetic anisotropy in Cu/Co/Cu(001) films as a function of the Cu overlayer thickness. Therefore, another factor which may be used to control the magnetocrystalline anisotropy is the film thickness. In this paper, we report on an oscillatory magnetocrystalline anisotropy with the magnetic layer thickness predicted for bcc Fe_n/fcc Au_5 (001) superlattices.

In this work, we consider bcc Fe_{2m-1}/fcc Au_5 (001) superlattices ($m = 1$–12). These superlattices have a simple tetragonal symmetry. Since the (001) surface unit cells of fcc Au

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and bcc Fe are lattice matched to within 1%, the two (bcc Fe and fcc Au) sublattices were assumed to have a common in-plane lattice constant (a for fcc Au). An ideal Fe/Au interface was initially assumed (but see the text later). The experimental fcc Au lattice constant ($a_{\rm fcc}^{\rm Au}$) was used to determine a and hence also the Fe and Au atomic sphere radii. The atomic sphere radii used for Fe and Au and the in-plane lattice constant are, respectively, 1.42, 1.59 and 2.88 Å.

We first performed all-electron self-consistent electronic structure calculations for the systems considered here by using the spin-polarized relativistic linear muffin-tin orbital (SPR-LMTO) method [6, 7]. The local density exchange–correlation potential parametrized by Vosko, Wilk and Nusair [8] was used. In the self-consistent calculations, the magnetization was assumed to be perpendicular to the Fe monolayer planes (i.e., $\boldsymbol{m} \parallel [001]$). We then calculated the spin-polarized relativistic band structure only once for the [001] and [100] magnetization orientations for these systems using the self-consistent potentials. We obtained the magnetocrystalline anisotropy properties via the so-called force theorem, i.e., we defined the anisotropy energy as the difference between the eigenvalue sums of these spin-polarized relativistic band structures for the two different magnetization directions concerned. In all of the present calculations, the basis functions used were s, p and d MTOs. The combined correction terms, which improve on the approximations made in the LMTO method [6], were included. The analytic tetrahedron technique was used to perform the Brillouin-zone (BZ) integrations [9]. In the self-consistent calculations, the k-mesh over the irreducible wedge (IW) of the BZ used was obtained by dividing the BZIW edge along the Γ–X direction into ten intervals. The resultant number of k-points over the BZIW is, for example, 198 (over 1/16 of the BZ) for bcc Fe_5/fcc Au_5 (001). For the magnetocrystalline anisotropy energy calculations, denser k-meshes in the IW of the BZ are necessary. The BZIW edge along the Γ–X axis was then divided into 20 intervals. The resultant number of k-points over the BZIW used is, e.g., 2205 (over 1/8 of the BZ) for bcc Fe_5/fcc Au_5 (001). The calculated anisotropy energy and moment are found to be converged within 10% with respect to the number of the k-points used.

Figure 1 displays the calculated magnetocrystalline anisotropy energy ($\Delta E_b = E_b^{100} - E_b^{001}$) as a function of Fe slab thickness n in bcc Fe_n/fcc Au_5 (001). A positive MAE means that the MAE 'prefers' the perpendicular magnetization whilst an in-plane magnetization would be preferred if the MAE is negative. Here we neglect the tiny in-plane magnetocrystalline anisotropy. The most remarkable feature is that the MAE oscillates in both magnitude and sign as the Fe slab thickness increases. The period of this oscillation is about ten monolayers (ML). Oscillatory magnetocrystalline anisotropies as a function of magnetic layer thickness have been found in recent tight-binding model calculations [10–13]. In particular, Cinal *et al* [11] reported that the anisotropy constant for the free-standing (001) bcc Fe slab oscillates as a function of the Fe slab layer thickness with a similar period. Nevertheless, the oscillatory anisotropy constant for the bcc Fe(001) slab obtained by Cinal *et al* [11] did not show any sign change and, furthermore, the anisotropy values were about one order of magnitude larger than the values presented here. The differences between the anisotropy energies reported earlier [11] and those presented in the present paper are attributed mostly to the differences between the previous semiempirical tight-binding approach and the present *ab initio* calculations, rather than to the effects of the Au layers. Interestingly, there is only one paper reporting *ab initio* theoretical evidence for the presence of the oscillatory magnetocrystalline anisotropy (including the sign change) in a magnetic multilayer [14].

First-principles calculations of the MAE in Au/Fe/Au (001) trilayers versus the Fe slab thickness have been reported before by Szunyogh *et al* [15] who found a small oscillation superimposed on a large perpendicular MAE. However, in reference [15] an unrealistic fcc structure with the Au lattice constant was assumed for the Fe slab, and this perhaps explains the

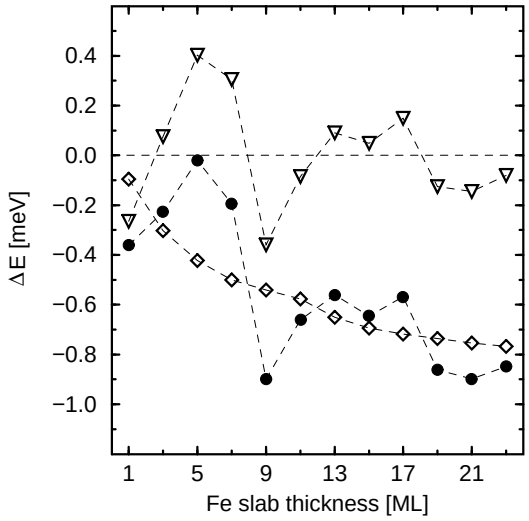

Figure 1. Magnetic anisotropy energies ($\Delta E = E^{100} - E^{001}$) for bcc Fe_n/fcc Au_5 (001) as a function of Fe slab thickness n (ML). Magnetocrystalline anisotropy energy (ΔE_b): triangles; magnetic dipole anisotropy energy (ΔE_d): diamonds; total anisotropy energy ($\Delta E_t = \Delta E_b + \Delta E_d$): circles. The lines are to guide the eye only. All of the anisotropies (including those plotted in figures 2 and 3 below) are per unit cell of the superlattice.

striking differences in the calculated MAE between the present work and previous calculations [15]. Another interesting difference is that for a single Fe monolayer, the perpendicular anisotropy was predicted in the previous calculations [15] whilst in the present work an in-plane magnetocrystalline anisotropy is found. The results of reference [15] nevertheless agree with our own previous calculations [16]. The difference again is mainly caused by the fact that a fcc structure was assumed in both previous calculations [15, 16]. The distance between interfacial Fe and Au monolayers is 1.74 Å in a bcc Fe/fcc Au (001) multilayer, and is 2.04 Å in a fcc Fe/fcc Au (001) multilayer. In this work, the MAE and other magnetic properties of Fe_1/fcc Au (001) versus the Fe–Au interlayer distance (d) were also calculated and are shown in figure 2. Clearly, the MAE as well as the Fe orbital magnetic moment in Fe_1/fcc Au_5 (001) are especially sensitive to d. In particular, figure 2 shows that as d increases, the magnetization will transform from an in-plane orientation to the perpendicular orientation at $d = 1.84$ Å. This interesting result is qualitatively consistent with the previous work of Strange *et al* [17] who predicted a flip in the easy axis of magnetization in tetragonally deformed bcc Fe as a function of axial ratio (c/a).

An experiment measures the total magnetic anisotropy energy of a magnetic system. The total anisotropy energy (ΔE) consists of the magnetocrystalline anisotropy energy (ΔE_b) discussed above and the magnetic dipole anisotropy energy (ΔE_d). Therefore, for comparison with experiments, one must know the magnetic dipole anisotropy energy as well. Also displayed in figure 1 are the calculated dipole anisotropy energy and the total anisotropy energy for the bcc Fe_n/fcc Au (001) superlattices. The dipole anisotropy energy of these Fe/Au superlattices was evaluated using the calculated magnetic moments by exploiting the Ewald-type lattice summation technique (see, e.g., reference [16] for more details). Note that unlike in the case of a single ferromagnetic slab where the dipole anisotropy energy is proportional to the film thickness [11, 15], the dipole anisotropy energy per unit cell for a superlattice is in general not proportional to the magnetic layer thickness especially when the

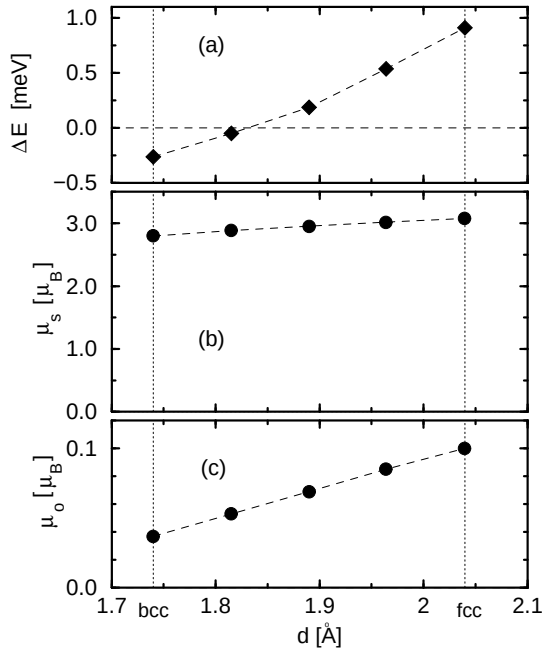

Figure 2. The magnetocrystalline anisotropy energy ΔE (a), Fe spin magnetic moment μ_s (b) and Fe orbital magnetic moment μ_o (c) in an Fe_1/fcc Au_5 (001) superlattice as a function of Fe–Au interlayer distance d. Vertical dotted lines at 1.74 and 2.04 Å, respectively, denote the Fe–Au interlayer distances for bcc and fcc Fe sublattices. The magnetic moments shown in (b) and (c) are for the perpendicular magnetization. The dashed curves are to guide the eye only.

magnetic layer becomes thicker than the nonmagnetic layer as shown in figure 1. Clearly, the total anisotropy energy 'prefers' an in-plane magnetization for the Fe slab thickness range considered here. This results from the large magnetic dipole anisotropy which always 'prefers' an in-plane magnetization. Experimentally, both the perpendicular and in-plane anisotropies have been reported for an ultrathin Fe_n/Au (001) film ($n \leqslant 3$), depending on the sample preparation conditions [18–20]. This is perhaps not surprising given the fact that figure 2 shows a sensitive dependence of the MAE on the Fe–Au interlayer distance. Furthermore, the calculated total energies suggest that the minimal energy $d \approx 1.85$ Å is close to the zero value of ΔE_b (see figure 2). Highly precise full-potential linear augmented-plane-wave calculations [21] suggested that in a Fe/Au (001) multilayer, the Fe–Au interlayer spacing should be very close to the average of the corresponding interlayer distances in bcc Fe and fcc Au (i.e., 1.74 Å). For thicker Fe slabs, an in-plane magnetization was always found in the previous experiments on Fe/Au multilayers [18–20]. To verify experimentally the predicted oscillatory behaviour of ΔE_b, further quantitative measurements of the MAE on high-quality Fe/Au multilayers are needed.

The oscillatory behaviour of the MAE found here is reminiscent of the oscillatory exchange coupling between two ferromagnetic layers separated by a paramagnetic metallic spacer layer [22]. It is well known that the oscillatory interlayer exchange coupling is mainly caused by the presence of the quantum well states in the spacer layer [23–26]. Indeed, Cinal *et al* [11] have very recently demonstrated that the oscillations of magnetocrystalline anisotropy with the Pd overlayer thickness that they found in (001) fcc Pd/Co/Pd slabs are associated with pairs of the quantum well states confined mainly in the Pd overlayers. Likewise, one may

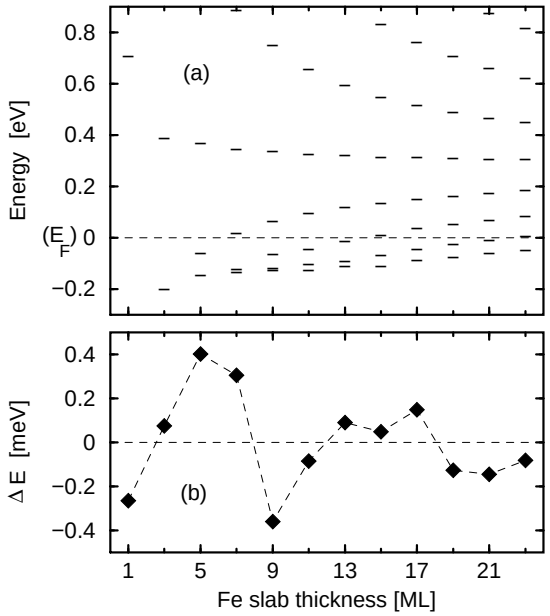

Figure 3. (a) The energy levels of the minority-spin Fe $d_{yz,zx}$-dominated states at the Brillouin zone centre in bcc Fe_n/fcc Au_5 (001) versus Fe slab thickness n (ML). The dashed line at 0 eV denotes the Fermi level. (b) For comparison, the magnetocrystalline anisotropy energy ΔE (figure 1) is replotted.

also speculate that the quantum well states in a magnetic layer drive an oscillatory MAE in a magnetic multilayer. Indeed, quantum well states have been observed in Fe/Au (001) multilayers [27–29]. In bulk bcc Fe, the majority- and minority-spin d bands are split and, as a result, the minority-spin d band is shifted above both the majority-spin Fe d band and the bulk Au d band [29]. Thus, in bcc Fe_n/fcc Au superlattices, the minority-spin Fe d states may form quantum well states in the vicinity of the Fermi level because of the band offset between Au d bands and minority-spin Fe d bands [27–29]. Here, inspection of the calculated site- and orbital-decomposed densities of states of the scalar-relativistic [30] band structures show that all of the minority-spin Fe d states except d_z^2 are confined in the Fe slab in the superlattices. Fe d_z^2 states can hybridize with Au sp bands and thus are rather delocalized along [001]. In figure 3(a), we plot the energy levels of the doubly degenerate Fe $d_{yz,zx}(\Delta_5)$-derived electronic states at the Γ point in the Brillouin zone as a function of Fe slab thickness. Interestingly, the energy levels of these states cross the Fermi level regularly with varying Fe slab thickness, and the crossing of the Fermi level by these states occurs roughly in the Fe thickness region where the MAE is positive (see figure 3(b)). Also note that the interval between two crossings (about 8–10 ML) is of the same order of magnitude as the period of the observed quantum well state at 2 eV above the Fermi level (14 ML) [27, 28]. Further band-structure calculations for bcc Fe_5/fcc Au_m ($m = 3, 7$) show that the energy levels of these states do not vary with Au slab thickness (within a few meV), confirming that the states are confined in the Fe slab. All of this points to the Fe $d_{yz,zx}$-derived quantum well states as the origin of the oscillatory MAE. When the spin–orbit coupling is included, the doubly degenerate Fe $d_{yz,zx}$-dominated states split for certain magnetization directions. It is found that the size of the splitting for the perpendicular magnetization is generally an order of magnitude larger than that for an in-plane magnetization (say, [100]) (e.g., 65 meV versus 5 meV). It has been demonstrated recently

that the contribution to the MAE due to the lifting of these degeneracies can become important when these states are in the close vicinity of the Fermi level [13]. Therefore, one could explain the oscillatory MAE found here as follows. When the Fe d-band quantum well states are away from the Fermi level, bcc Fe/fcc Au (001) superlattices have a negative (in-plane) MAE. When these quantum well states approach the Fermi level, their contributions to the MAE become dominant and hence the superlattices show a perpendicular magnetocrystalline anisotropy. Nevertheless, definite conclusions cannot be reached until further tedious numerical analyses of the contributions from the electronic states over the entire Brillouin zone are carried out.

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