**Ab initio** prediction of giant magnetostriction and spin-reorientation in strained Au/FeCo/MgO heterostructure

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**ABSTRACT**

Employing *ab initio* electronic structure calculations we have investigated the magnetostrictive properties and the effect of epitaxial strain on the magnetic anisotropy (MA) of Au/FeCo/MgO heterostructure. Under small expansive strain on the FeCo layer the system exhibits an in-plane MA. The calculations reveal that the strain dependence of the MA is nonlinear and that the FeCo film undergoes a spin reorientation at a critical strain between 2 and 4%. The underlying mechanism is the strain-induced shift of the spin–orbit coupled d-states of the Fe atoms. We predict a giant magnetostriction coefficient of about $420 \times 10^{-6}$ in the heterostructure.

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**1. Introduction**

Magnetic nano-junctions of heavy-metal/ferromagnet/insulator (HM/FM/I) are of great interest in spintronic devices such as high density and nonvolatile magnetic random access memory (MRAM). The magnetization reversal is induced by spin-polarized current via the spin transfer torque (STT-RAM) [1] or by an electric field via the magnetoelectric effect (MeRAM) [2–4]. Au/FeCo/MgO is a promising system since recent experiments by Shiota et al. showed voltage-induced switching of the magnetic easy axis from in- to out-of-plane direction [2].

Since there is often a fairly large lattice mismatch (4–5%) between the I, the FM and HM layers, the component layers are under large uniaxial strain which can in turn modify significantly the magnetic properties of the heterostructure including the magnetic anisotropy (MA) which arises from the spin orbit coupling (SOC). *Ab initio* calculations have recently investigated the effect of hydrostatic pressure on the MA of bulk FeCo alloys [5,6].

In this work we report results of *ab initio* electronic structure calculations of the effect of epitaxial strain on the MA of the Au/FeCo/MgO (001) heterostructure. We find that the strain dependence of MA is nonlinear and that the system undergoes a spin reorientation transition at a critical strain between 2 and 4%. The underlying mechanism is the strain-induced shift of spin–orbit coupled d-states of the interfacial Fe ions. We also predict that the strain dependence of the MA yields a giant magnetostriction coefficient of 420 ppm (parts per million).

**2. Computational method and model**

First-principles density functional calculations were performed within the framework of the plane wave projector augmented wave formalism [7], as implemented in the Vienna *ab initio* simulation package (VASP) code [8–10]. The generalized gradient approximation [11] has been employed to treat the exchange-correlation functional. To simulate the epitaxial growth of the Au/FeCo/MgO junction, we employed a slab supercell along the [001] direction consisting of three monolayers (MLs) of Au, three MLs of B2-type FeCo, seven MLs of rock-salt MgO and a 15 Å-thick vacuum region separating the periodic slabs [Fig. 1(a)]. The FeCo/MgO interface is modeled by placing the O atoms atop the Fe atoms [Fig. 1(b)]. The interfacial Fe atoms at the Fe/MgO and Fe/Ta interfaces are denoted by Fe1 and Fe2 atoms, respectively.

We build up supercells by aligning the $\langle 110 \rangle$ axis of MgO and Au with the $\langle 100 \rangle$ axis of FeCo. The FeCo layer is under large expansive strain, $\eta_{\text{FeCo}}$, of about 4% assuming that the MgO substrate is unstrained. On the other hand, recent experiments showed that ultrathin (~2 nm) MgO film is significantly compressed to match the lattice constant of the FM film [12]. In order to study the effect of lateral strain under different growth conditions, we have varied $\eta_{\text{FeCo}}$ from zero to 4%. At each strain, the magnetic, electronic...
degrees of freedom and atomic $z$ positions are fully optimized until the atomic forces become less than $5 \times 10^{-3}$ eVÅ and the change in the total energy between two ionic relaxation steps is smaller than $10^{-6}$ eV. The plane-wave cutoff energy is 500 eV and the Monkhorst–Pack $k$-mesh for the relaxation calculations was $15 \times 15 \times 1$. The dipole correction to the total energy is applied [13]. The valence electrons are treated in the scalar relativistic approximation and the SOC of the valence electrons is included using the second-variation method [14] employing the scalar-relativistic eigenfunctions of the valence states. Employing a $31 \times 31 \times 1$ $k$-point mesh, the MA per unit interfacial area, $A$, is determined from $MA = \left[ E_{\parallel 100} - E_{001} \right] / A$, where $E_{\parallel 100}$ and $E_{001}$ are the total energies with magnetization along the [100] and [001] directions, respectively.

3. Results and discussion

The interlayer atomic distances, spin moment and orbital-moment difference of Fe1, Fe2, and Au1, and MA of the system under various strains on the FeCo are listed in Table 1. At $\eta_{FeCo} = 0$, the interlayer distances in the Au cap are larger than those in the FeCo layer. This is due to the large compressive strain on the Au layer, since the lattice constant of bulk Au is larger than that of the B2-type FeCo. In the FM layer, the interlayer Co–Fe1 distance is significantly smaller than that of Fe2–Co due to the displacement of the Co atom. As expected, these distances decrease with increasing expansive strain. The magnitude of spin moments of Fe1 and Fe2 are comparable and vary by less than 1% with strain. Au ions have negligible spin moment, even for the Au1 next to the FM layer. On the other hand, the orbital moment difference $\Delta m_o = m_{O}^{(001)} - m_{O}^{(100)}$ of Fe1 is at least 3 times as large as that of Fe2 and exhibits strong strain dependence. Interestingly, a magnetic switching from in-plane to out-of-plane direction occurs with increasing strain in the range from 2% to 4%. Note that the strain variation of MA is nonlinear and correlates to the change of $\Delta m_o$ of Fe1.

In order to understand the role of the MgO and the Au cap on the MA we have carried out calculations of the MA of the free standing 3 ML FeCo film (vacuum/FeCo/vacuum) and the MgO/FeCo/vacuum for $\eta_{FeCo} = 0, 4\%$. Under zero stain the MA of the free standing and MgO/FeCo/vacuum systems are 1.28 and 2.70 erg/cm$^2$, respectively, indicating an out-of-plane magnetization orientation in sharp contrast to the Au/FeCo/MgO heterostructure which favors in-plane magnetization. At 4% the MA of the free standing and MgO/FeCo/vacuum systems are $-0.31$ and $-0.68$ erg/cm$^2$, respectively, indicating that both systems undergo a transition from out-of-plane to in-plane direction with increasing strain. These results show that the MgO/FeCo interface provides a large contribution to the perpendicular MA at zero strain which arises from the Fe 3d–O 2p hybridization. It is also interesting to note that the MA of the MgO/FeCo/vacuum system is more sensitive to strain compared that of the free standing FeCo film. This is due to the sensitivity of the Fe–O bond length and hence the hybridization to strain. The opposite sign of MA for the Au/FeCo/MgO system compared to the corresponding uncapped systems under the same strain is clearly induced by the Au capping layer. The underlying mechanism is the shift in the energy levels of Fe d-states and the changes in energy- and $k$-resolved distribution of the Fe d-orbital characters, which are induced by the capping layer.

To elucidate the mechanism of the strain effect, we show in Fig. 2 the evolution of the minority-spin Fe d-derived bands close to the Fermi energy along a segment of the $\Gamma$–$\Xi$($\Sigma$) direction with increasing strain $\eta_{FeCo} = 0, 2, 4\%$. Within second-order perturbation of the total energy due to SOC constant, $\xi$, the MA is determined by the matrix elements of the orbital angular momentum operators $\hat{L}_x$ and $\hat{L}_z$ between occupied and unoccupied d-states and by the energy difference between these states. Since the Fe-derived majority spin states are well below the Fermi energy, the MA contribution from the SOC between states of opposite spin can be neglected. Thus, the MA can be expressed as [15]

$$ MA \propto \xi^2 \sum_{\alpha, \beta} \left[ \left( \psi^\alpha_\uparrow(E^\alpha_{\uparrow}) \right)^2 - \left( \psi^\alpha_\downarrow(E^\alpha_{\downarrow}) \right)^2 \right] \frac{E^\alpha_{\downarrow} - E^\alpha_{\uparrow}}{E^\alpha_{\uparrow}}. $$

where $\psi^\alpha_\uparrow(E^\alpha_{\uparrow})$ and $\psi^\alpha_\downarrow(E^\alpha_{\downarrow})$ are occupied and unoccupied minority-spin states (energies), respectively; and $\hat{L}_{x,z}$ is the $x$ ($z$) component of orbital angular momentum operator.

At zero strain, the Fe1 $d_{xy,z}$-derived states are mainly unoccupied and couple through $\hat{L}_x$ to the occupied Fe1 $d_{x^2-y^2}$ – (green curves below the Fermi level) and $d_{xy}$-derived states (green circles near $\Gamma$) yielding negative MA. At 2% strain, the occupied

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**Table 1**

<table>
<thead>
<tr>
<th>Strain(%)</th>
<th>$d_{Fe-Co}$ (Å)</th>
<th>$m_o$ ($\mu_0$)</th>
<th>$\Delta m_o(10^{-2} \mu_0)$</th>
<th>MA (erg/cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>2.191</td>
<td>2.795</td>
<td>0.046</td>
<td>2.1</td>
</tr>
<tr>
<td>2</td>
<td>2.123</td>
<td>2.808</td>
<td>0.048</td>
<td>2.1</td>
</tr>
<tr>
<td>4</td>
<td>2.056</td>
<td>2.773</td>
<td>0.048</td>
<td>3.2</td>
</tr>
</tbody>
</table>

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\( \Delta V = 2e_1 + e_3 + 2e_1e_3 + e_1^2 = \eta_{\text{FeCo}} - \eta_{\text{FeCo}}^2. \)  

Thus, Eq. (2) can be rewritten in the form

\[
E(e_i) = \frac{E_0}{V} - P(V)\eta_{\text{FeCo}} + \left[ P(V) + G_1 \right] \eta_{\text{FeCo}}^2.
\]  

Fig. 3 shows the \textit{ab initio} calculated energy density of Au/FeCo/MgO as a function of \( \eta_{\text{FeCo}} \). By fitting the data to Eq. (6), we find that \( P(V) = 20.8 \text{ GPa} \) and \( C_{11} = 251.8 \text{ GPa} \). For the bulk Fe\(_{50}\)Co\(_{50}\) system, the experimental value of \( C_{11} \) is 244 GPa (inferred from \( C = (C_{11} - C_{12})/2 = 61 \text{ GPa} \)) [19].

The strain dependence of the \textit{ab initio} calculated MA can be fitted to the magnetoelastic equation [20]:

\[
MA = K_f^f + B_{11}^f \left( \frac{2C_{11} + D_{11}}{(G_1 + D_{11})^2} \right)
\]

\[
+ B_{11}^f \left( 1 + \frac{C_{11}^2}{(G_1 + D_{11})^2} \right) \eta_{\text{FeCo}}
\]

\[
+ \frac{D_{11}\eta_{\text{FeCo}}^2}{2 \left( \frac{C_{11}}{G_1 + D_{11}} \right)}.
\]

Here, \( K_f^f \) is the effective interface magnetocrystalline anisotropy, \( t \) is the FM thin film thickness, and \( B_{11} \) and \( D_{11} \) are the first and second order magnetoelastic coefficients, respectively. Using the calculated \( C_{11} = 251.8 \text{ GPa} \), the FM film thickness \( t = 2.677 \text{ Å} \), and \( B_{11} = 6.0 \times 10^6 \text{ erg/cm}^2 \) and \( D_{11} = 44.8 \times 10^6 \text{ erg/cm}^2 \), and \( K_f^f = -0.61 \text{ erg/cm}^2 \). For comparison, for bulk Fe\(_{50}\)Co\(_{50}\), the experimental value of the first-order magnetoelastic coefficient is \( B_{11} = -3 \times 10^6 \text{ erg/cm}^2 \) [19].

The magnetostriction coefficient can be determined from [21]

\[
\lambda_{001} = -\frac{2B_{11}}{3(G_1 - G_2)}.
\]

Using the calculated values above for \( B_{11}, C_{11}, \) and \( G_2 = G_{11}/2, \)
we find that $\lambda_{001} = 424 \times 10^{-6}$. This value is one-third of that estimated for the Fe$_{34}$Co$_{66}$ thin film ($\lambda_{001} = 1300 \times 10^{-6}$) under the assumption of vanishing $\lambda_{111}$ [22], and is of the same order of magnitude as the giant values reported for Tb-based thin films [23].

4. Conclusion

In summary, we have presented \textit{ab initio} electronic structure calculations that investigate the effect of epitaxial strain on the MA of the Au/FeCo/MgO epitaxial heterostructure. At zero strain on the FM layer the junction exhibits in-plane MA due to the large $\langle x^2 - y^2 \rangle \tilde{L}_4(i(xz, yz))$ and $\langle xy \tilde{L}_4(i(xz, yz))$ spin–orbit coupling of the Fe atom at the FeCo/MgO interface. The strain dependence of MA is nonlinear. We predict magnetic switching of the magnetic easy axis from in- to out-of-plane orientation at a critical strain between 2 and 4%. The underlying mechanism lies on the strain-induced shift of the spin–orbit coupled $d$-states of the interfacial Fe atom. We predict a giant magnetostriction coefficient of 424 ppm in the heterostructure. These results demonstrate that strain engineering can open an avenue to tailor the magnetic properties for spintronic applications.

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References

[20] Derivation of this magnetoelastic equation and complete MA data for fitting to be published elsewhere.