Short Communication

Ab-initio Determination of Magnetic Interface Coupling Constants for Magnetic Multilayers

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(Received 21 July 1997, received in final form 30 July 1997, accepted 8 September 1997)

PACS.75.70.Cn – Interfacial magnetic properties (multilayers, magnetic quantum wells, superlattices, magnetic heterostructures

PACS.71. – Electronic structure

Abstract. —The magnetic (in-plane) interface coupling energy of an Au(100)/FeAu$_3$Fe/Au(100) multilayer system has been calculated using the well-known fully relativistic spin-polarized Screened KKR method. The coupling energy was expanded in polynomials of $\cos(\delta)$ in order to compare it with calculations using the Force Theorem method prescription. The second order term in the polynomial expansion is important when looking at total energy differences. The intention of this paper is to show the numerical feasibility of using the force theorem on particular model systems. In another paper we apply it to existing physical systems.

The discovery of the oscillatory behavior of magnetic interface interactions especially in the asymptotic limit has been the subject of several studies [1]. In these papers the problem of interface magnetism (*i.e.*, the question of the so-called in-plane magnetic interface coupling energy of magnetic multilayer systems) is dealt with extensively.

Suppose such a multilayer system consists of two monolayers of Fe, separated by m layers of a non-magnetic spacer, and is sandwiched semi-infinitely by the spacer material. By considering only in-plane orientations of the magnetization in the two Fe planes, only the relative angle of these two orientations should matter. The magnetic interface coupling energy,

$$E_{\mathrm{a}}(\delta) = E(\delta) - E(0), \quad 0 \leq \delta \leq \pi, \tag{1}$$

namely the energy difference between a parallel configuration and an arbitrary (relative) in-plane configuration of the magnetization in the Fe planes, is usually assumed to be proportional

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to $(1 - \cos(\delta))$, a functional dependence inherent also to the so-called torque method [2], since the torque $T(\delta)$ is defined by the derivative of $E_a(\delta)$ with respect to δ, $T(\delta) = -\partial E_a(\delta)/\partial\delta$.

When the spacer thickness m is varied, $E_a(\delta)$ oscillates with respect to m and it is precisely these oscillations which are of interest. In principle $E_a(\delta)$ contains higher order terms in $\cos(\delta)$. By defining the magnetic interface coupling energy $E_a(\delta)$ as

$$E_a(\delta) = \sum_n E_n(\delta), \quad E_n(\delta) = a_n \left(1 - \cos^n(\delta)\right), \tag{2}$$

the question arises whether the constant a_2 can be determined from *ab-initio* calculations, but also whether even higher order terms are significant. It should be noted that in the context of interface exchange coupling, the coefficients a_1 and a_2 are usually referred to as the bilinear and the biquadratic exchange coupling coefficients (see also [3, 4]).

In order to address this problem — as a case study — the system $Au(100)/Au_2FeAu_3FeAu_2/Au(100)$ is investigated, using the spin-polarized (fully) relativistic version of the Screened KKR method [5–7], whereby — as indicated — two layers of Au serve as a buffer to the semi-infinite substrate (for technical and numerical details see in particular Refs. [7, 8]). All calculations reported here are based on the density functional as given in [9]. For the nine atomic layers in the intermediate region, namely for the layer sequence {Au, Au, Fe, Au, Au, Au, Fe, Au, Au}, the relative angle δ in the magnetic configurations

$$C(\delta) = \{0, 0, 0, 0, 0, \delta, \delta, \delta, \delta\} \tag{3}$$

was varied in steps of $n\pi/4, n = 0, 1, \ldots, 4$, whereby for $\delta = 0$ (ferromagnetic configuration) the orientation of the magnetization was chosen to point along the x-axis. In one particular case, namely for $\delta = \pi/2$, also a configuration

$$C'(\delta) = \{0, 0, 0, 0, \delta/2, \delta, \delta, \delta, \delta\}, \tag{4}$$

because of its symmetrical arrangement was investigated.

In what follows let $\epsilon_a(\delta)$ denote the *actually calculated* magnetic interface coupling energy at a particular value of δ,

$$\epsilon_a(\delta) = \epsilon(\delta) - \epsilon(0). \tag{5}$$

where $\epsilon(\delta)$ refers to the total energy of a particular configuration $C(\delta)$. Quite clearly the constants a_1 and a_2 in (2) can then be determined using the relations

$$a_1 = \frac{1}{2}\epsilon_a(\pi), \quad a_2 = \epsilon_a(\pi/2) - \frac{1}{2}\epsilon_a(\pi), \tag{6}$$

while higher order terms in $\cos(\delta)$ result from inspecting the following differences:

$$\Delta E_a(\delta) = \epsilon_a(\delta) - [E_1(\delta) + E_2(\delta)], \tag{7}$$

where — as should be recalled — $E_n(\delta)$ is defined in (2).

The magnetic interface coupling energies $\epsilon_a(\delta)$ were also calculated using the so-called Force Theorem (FT), which very successfully was applied and able to predict the perpendicular magnetic anisotropy energy for Au(100)/Fe [7, 8] and Au(111)/Co [10] multilayers. It should be noted that within the Force Theorem only one magnetic configuration, namely the ferromagnetic one ($\delta = 0$), is calculated selfconsistently, and the magnetic interface interaction energy is obtained from the following band energy differences, see in particular [8],

$$\epsilon_a(\delta) \approx \epsilon_{band}(\delta) - \epsilon_{band}(0). \tag{8}$$

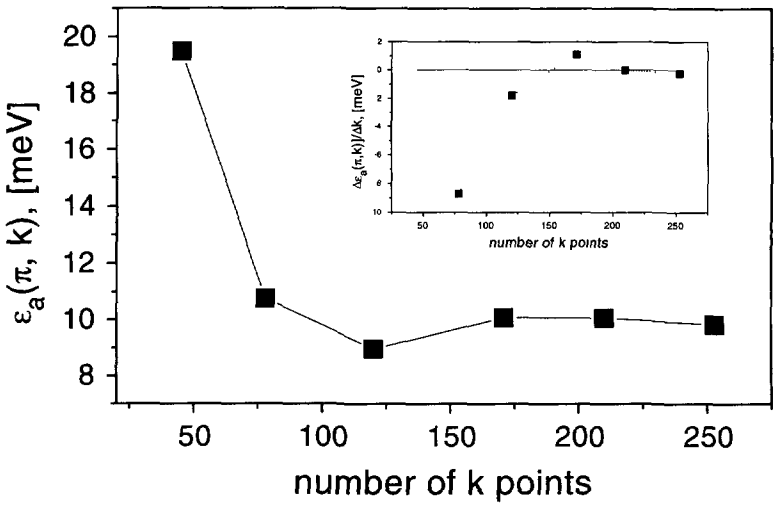

Fig. 1. — Convergence of $\epsilon_a(\pi)$ with respect to the number of k-points in the irreducible part of the Surface Brillouin zone. The insert shows the numerical derivative of this curve with respect to k. See also [8].

Within the Force Theorem $\epsilon_{\text{band}}(\delta), \delta \neq 0$, refers therefore to the band energy of a magnetic configuration $C(\delta)$, calculated, however, using the layer dependent potentials from the $\delta = 0$ selfconsistent calculation.

In Figure 1 the convergence of $\epsilon_a(\pi)$ as calculated in terms of total energies is shown with respect to the number of k-points in the irreducible part of the Surface Brillouin Zone (SBZ) used to perform the necessary SBZ-integrations. It should be noted that for each entry in this curve two fully converged selfconsistent calculations are needed. As one can see from the insert in this figure, for more than 200 k-points, the magnetic interface coupling energy is converged to an accuracy of about 0.1 meV. In the following all total energy differences refer to one and the same k-mesh, namely 210 k-points.

Table I summarizes the results for the first two coefficients in (2) as based on the configurations defined in (3). These results show that when using the exact formulation, namely in terms of total energy differences, the first coefficient a_1 is larger by a factor of 4 than the one predicted by the FT. Confirming the usual experimental experience, in the total energy case the second coefficient a_2 is at best one order of magnitude smaller than the first one, while in the FT-case the ratio $|a_2|/a_1$ is much smaller.

Figures 2 and 3 illustrate the variation of $E_a(\delta)$ with respect to δ in these two cases. From Figure 2 it is evident that at least in the total energy case in (2) an expansion up to $n = 2$

Table I. — *Fitting parameters [meV].*

	total energy	force theorem
a_1	5.027	1.359
a_2	−0.687 (−0.551)	0.034
$\lvert a_2\rvert/a_1$	0.137 (0.109)	0.025

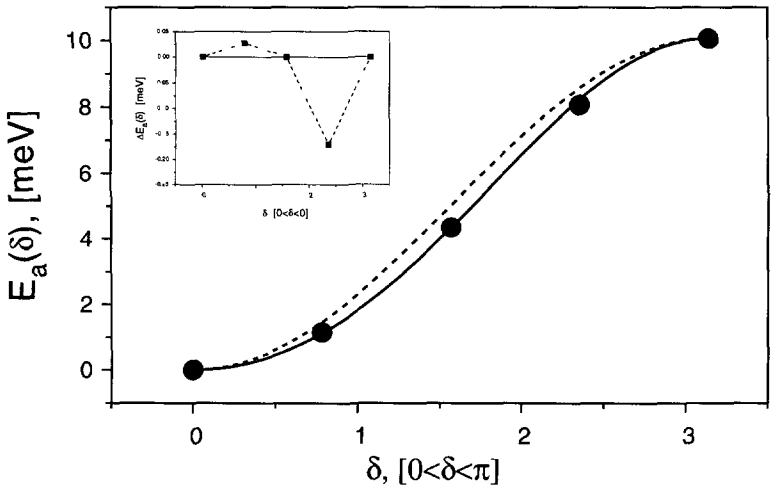

Fig. 2. — Fit of the magnetic interface coupling energy $E_a(\delta)$ to total energy calculations corresponding to configurations $C(\delta)$. The calculated values of $\epsilon_a(\delta)$ are shown as full circles, $(E_1(\delta) + E_2(\delta))$ as full line and $E_1(\delta)$ as dashed line. The insert shows $\Delta E_a(\delta)$, see the definitions (2)-(8).

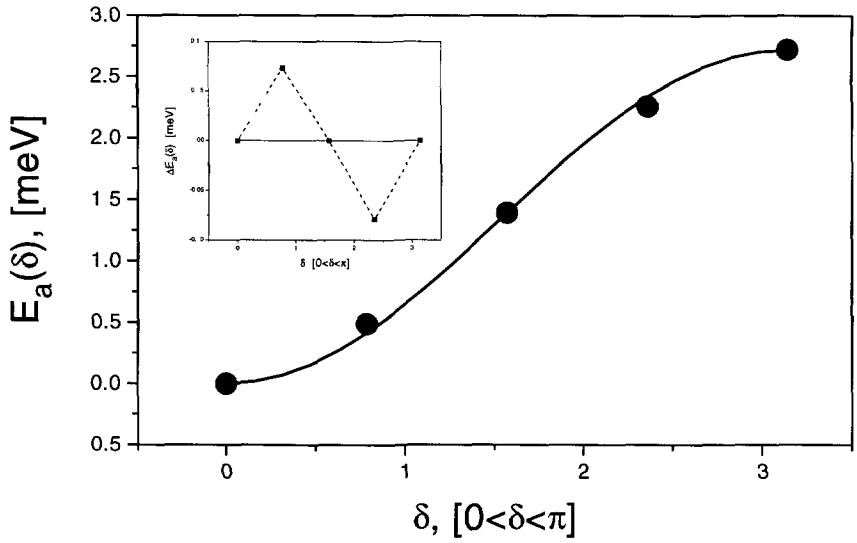

Fig. 3. — Fit of the magnetic interface coupling energy $E_a(\delta)$ to force theorem calculations. The calculated values of $\epsilon_a(\delta)$ are shown as full circles, $(E_1(\delta) + E_2(\delta))$ as full line. $E_1(\delta)$ falls on top of the full line. The insert shows $\Delta E_a(\delta)$, see the definitions (2)-(8).

is needed to fit the calculated values, while, as can be seen from Figure 3, as well as from Table I for the FT-values, a fit with $n = 1$ seems to be reasonably sufficient.

In order to illustrate the effect of selfconsistency, in Figure 4 the following differences of the layer-resolved magnetic moments,

$$\Delta m(\delta) = m(\delta) - m(0), \tag{9}$$

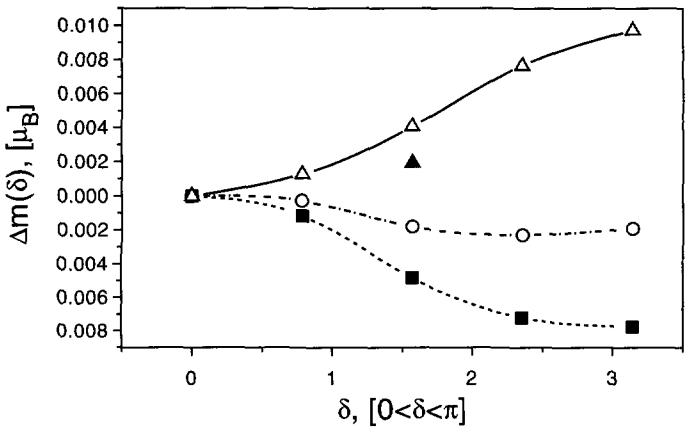

Fig. 4. — Layer-resolved magnetic moment differences $\Delta m(\delta)$ (9). 4 cases: (open triangles) center Au-spacer layer, using configuration $C(\delta)$; (full triangle) center Au-spacer layer, using configuration $C'(\delta)$, $\delta = \pi/2$; (full squares) Au-spacer layers at the Fe/Au interface, using configuration $C(\delta)$; (open circles) Fe-layers, using configuration $C(\delta)$. The lines serve as guidance to the eye.

are displayed with respect to δ. As one can see for configurations of type $C(\delta)$ the Au difference moments vary continuously with δ, while the corresponding differences for the Fe layers (both Fe layers give virtually the same differences) vary only very little for $\pi/2 < \delta \leq \pi$. It is worthwhile to mention that for $\delta = \pi$ the moment in the center Au layer is exactly zero. In Figure 4 for $\delta = \pi/2$ also the case of the configuration $C'(\delta)$ is indicated. The corresponding magnetic interface coupling energy is by 0.14 meV smaller than the one referring to a configuration of type $C(\delta)$, leading in turn to the coefficient a_2 given in brackets in Table I.

Inspecting the inserts in Figures 2 and 3 it is obvious that even in the case of total energies the remainder $\Delta E_a(\delta)$ (7) is rather small and in value close to the convergence criterion for the respective magnetic interface coupling energies $\epsilon_a(\delta)$. Nevertheless, it is important to recall that the expression for $E_a(\delta)$ in (2) is based on the idea of a SU_2 rotation (see, *e.g.*, [4]), which of course is not quite correct in a fully relativistic spin-polarized approach case (see also the discussion in [11]), although the deviation from a point group operation at least in the present case seems to be of minor importance. The shape of $\Delta E_a(\delta)$ in Figure 2 might very well reflect also such a deviation.

Summary

It was shown that by using judiciously a total energy concept for the in-plane magnetic interface coupling energy, an accurate determination of the first two coefficients for an expansion of the magnetic interface coupling energy in terms of powers of $\cos(\delta)$ can be achieved. In contrast, at least for thin spacer systems, the otherwise very useful and practicable Force Theorem can not be expected to give a reasonable value for the ratio $|a_2|/a_1$. It should also be noted that the numerical accuracy of this method is such that it is equally accurate for small thin film systems as well as for large $n > 30$ multilayer systems and that these are not artificial size effects.

Acknowledgments

This paper resulted from a collaboration partially funded by the TMR network on "*Ab-initio* calculations of magnetic properties of surfaces, interfaces, and multilayers" (contract No.: EMRX-CT96-0089). Financial support was provided also by the Center of Computational Materials Science (GZ 308.941), the Austrian Science Foundation (P11626), and the Hungarian National Science Foundation (OTKA T24137). We also wish to thank the computing center IDRIS at Orsay as part of the calculations was performed on their Cray T3D machine.

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