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Published in:
Physical Review Letters

DOI:
10.1103/PhysRevLett.100.057205

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2008

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Download date: 23-04-2024
Spin Tunneling in Junctions with Disordered Ferromagnets


1Department of Applied Physics, cNM, Eindhoven University of Technology, The Netherlands
2ESM, IMM, Faculty of Science, Radboud University Nijmegen, The Netherlands
3STFC Daresbury Laboratory, Warrington, Cheshire, WA4 4AD, United Kingdom
4CEMES- CNRS, 29, rue Jeanne Marvig B.P. 94347 F-31055 Toulouse Cedex 4, France

(Rceived 4 October 2007; published 6 February 2008)

Right from its inception, experimental and theoretical endeavors in electron tunneling have been dedicated to the understanding of the role of the electrode and barrier electronic structure. For ferromagnetic films, one aspect of their electronic structure—the tunneling spin polarization (TSP)—was measured with superconducting tunneling spectroscopy (STS) by Meservey and Tedrow [1]. Although some preliminary effort was undertaken to study the role of the band structure of ferromagnetic films in tunneling [2], no definitive observations were made till the advent of tunnel magnetoresistance (TMR) in magnetic tunnel junctions (MTJs). Then, Yuasa et al. [3] and LeClair et al. [4] experimentally demonstrated the influence of epitaxial Fe and textured Co films on TMR and tunneling conductance, respectively. Regarding the nature of the electronic wave functions that govern the tunneling probability through AlO₃, the dominance of the spherically symmetric s-like electrons has been experimentally demonstrated [5,6]. Recently, spintronics has witnessed a rapid rise in the importance of amorphous ferromagnets like CoFeB. They have contributed to huge TMR in AlO₃ [7] and MgO [8] based MTJs. They have also been used to observe the novel spin-torque diode effect [9] and facilitated record-low switching currents in spin-torque based MTJs [10]. Although their emerging importance in spintronics is unquestionable, neither has there been a theoretical and experimental analysis of their atomic and electronic structure, nor has the impact of these properties on their TSP been investigated.

In this Letter, we explore the correlation between ferromagnet morphology, its electronic structure, and their combined impact on TSP. One unique aspect—crystallization of amorphous CoFeB with a single high temperature anneal (≥ 250 °C [11])—is exploited to study the structural, magnetic, and TSP related properties of amorphous and crystalline CoFeB in the same sample. Indeed, such control on morphology is not accessible in elemental magnetic films. The high temperature anneal stipulates a crucial requirement for our junctions; viz., the barrier properties should not change after annealing to ensure comparison between the TSP of as-deposited and annealed CoFeB. Contrary to alternative barriers like MgO, AlO₃ barriers are known to exhibit no TSP related changes after anneals up to Tₐ = 500 °C [12,13]. When the structure of Co₇₂Fe₂₈B₈ is intentionally transformed from amorphous to highly textured fcc, we notice that a correlated alteration of the CoFeB electronic structure is induced. Contrary to one’s primary intuition, this alteration of the electronic structure manifests in an intrinsically larger TSP for amorphous CoFeB as compared to that of highly textured fcc CoFeB. First-principles atomic structure calculations of amorphous CoFeB are found to be consistent with extended x-ray absorption fine structure (EXAFS) measurements. Remarkably, electronic structure calculations based on this atomic structure exhibit a conspicuous agreement between theory and experiment for a complex system [14], and the dominance of s electrons in tunneling through AlO₃ [5,6].

The inset in Fig. 1(a) shows a representative TSP measurement for an as-deposited 120 Å CoFeB film using STS [11]. Regardless of the CoFeB thickness (d), for as-deposited samples, we consistently measure a TSP above...
A clue to the probable reason behind this change in the TSP of thin CoFeB films can be found in x-ray diffraction (XRD) measurements on films of corresponding thickness. In Fig. 1(b), the grain size perpendicular to the film plane, calculated using the Paul Scherrer formula, and normalized to the film thickness, is plotted as a function of $T_a$. This plot indicates that, in progressively thinner films, the grain sizes become comparable to the film thickness after the anneal. For $T_a = 450 \, ^\circ \text{C}$ and $d = 120 \, \text{Å}$, the average grain size is almost equal to the film thickness, suggesting the presence of crystalline CoFeB at the interface with the AlO$_x$ barrier. This hypothesis is substantiated by high resolution transmission electron micrographs (HRTEM). Figure 1(c) shows a junction with a 700 Å CoFeB layer, while Fig. 1(d) corresponds to a 60 Å CoFeB layer, both annealed at 450 °C. For the 700 Å film, a close inspection of the barrier-ferromagnet interface region shows hardly any crystalline CoFeB at the interface, though we observe CoFeB crystallites in the bulk of the film (not shown). In sharp contrast, we observe almost comprehensive crystallization of CoFeB in the case of the 60 Å film, especially at the barrier-ferromagnet interface. Together, the XRD and HRTEM data strongly advocate that thicker films ($d \geq 500 \, \text{Å}$) do not crystallize completely after the anneal, especially at the interface with amorphous AlO$_x$, and consequently show a TSP similar to that of as-deposited amorphous CoFeB. On the contrary, thinner films crystallize virtually completely, and the TSP of crystalline CoFeB at its interface with AlO$_x$ manifests its intrinsic value. Note that the interface sensitivity of the TSP [14] is implicitly demonstrated within this inference. Furthermore, consistent with the observations of Takeuchi et al. [15], in crystalline films, the out-of-plane grain size is limited by the film thickness, while the in-plane grain size (150–200 Å) is similar to that observed in thicker films. As anticipated for such a Co rich composition, high angle XRD also confirms that CoFeB crystallizes in a highly (111) textured fcc structure.

Having established that the lowering of the CoFeB TSP is closely related to its crystallization, we embark on first-principles calculations using density functional theory within the generalized gradient approximation [16]. The self-consistent electronic structure and interatomic forces are calculated with the projector augmented wave method [17] using the Vienna ab initio molecular dynamics package (VASP) [18]. For reliable determination of the amorphous structure, the ensemble is heated above its melting point and equilibrated in the liquid state for time periods long enough to allow diffusion beyond one lattice spacing, and then rapidly quenched to form the amorphous state. Structural and electronic properties of two 108 atom ensembles are compared to three 54 atom ensembles for further verification and statistics. It is noteworthy that ensembles without B atoms do not quench in an amorphous structure, indicating the key role played by $\sim 7$ at.% B in rendering CoFeB amorphous. In the fcc case, the atoms are
randomly placed in nominal positions of an fcc lattice, and then allowed to relax. The total energy of the amorphous ensembles is invariably found to be higher than that of the distorted fcc ensembles, consistent with the fact that as-deposited amorphous films crystallize after an anneal.

Representative structures of one amorphous and one fcc ensemble are shown in Figs. 2(a) and 2(b) together with the partial radial distribution functions [pRDFs—Figs. 2(c) and 2(d)]. Irrespective of the size of the unit cell (108 or 54 atoms), the pRDFs show no significant difference in the inter- or intra-atomic coordination up to \( r = 5.5 \, \text{Å} \), indicating that a 108 atom ensemble is of sufficient size. To gain insight in the atomic structure of amorphous films, EXAFS measurements were performed on Co and Fe K edges. The measured and fitted data are shown in Fig. 2(e) and the corresponding Fourier transform (FT) in Fig. 2(f).

The oscillations seen in Fig. 2(e) are characteristic of disordered solids where usually the first coordination shell is the largest contributor to the fine structure, as is evident in the single peak dominating the FT. Keeping in mind the difficulties in fitting an amorphous structure, the fit to the second coordination shell around 3.5 Å is almost completely wiped out in comparison to the fcc case.

Considering the amorphous nature of the barrier, one might argue that \( k_{\|} \) conservation is highly unlikely in tunneling through AlOx. In the first instance, if one neglects any issue related to the barrier or interface electronic structure, the SP of \( s \)-like electrons, which have been experimentally shown [5,6] to dominate tunneling through AlOx, is the only quantity that needs consideration. Table I shows the calculated average \( s \)-electron SP at the Fermi level (\( E_F \)) for Co, Fe, and B in the amorphous and fcc case.

Assuming that the concentration at the interface is similar to that in the bulk, we obtain the alloy SP by weighting these individual SPs with their concentrations [1]. The last columns of Table I compare the measured TSP to the calculated alloy SP. For both the amorphous and fcc case, the calculated SPs of 50% \( \pm 0.2\% \) and 41% \( \pm 0.5\% \) are in surprisingly good agreement with the measured TSPs of 53% \( \pm 0.5\% \) and 44% \( \pm 0.5\% \), respectively. Most strikingly, the difference of \( \sim 9\% \) between the two measured TSP values is directly reflected in the calculations as well, indicating that this difference may arise from the disparity in the band structure of bulk amorphous and fcc CoFeB. It is noteworthy that the values of the element-specific and the alloy SPs are remarkably similar from one unit cell to another for the 5 amorphous and 2 fcc unit cells studied.

The errors in Table I are deduced from the variations in the first- and second-nearest neighbors of the calculated alloy SP. Inset in (d) shows pRDF for Co-B.

Interface bonding effects have been calculated to have pronounced effects on the TSP [20]. However, given the amorphous nature of AlOx, these are rather difficult to predict, and in reality, they are an average over the con-
TABLE I. Calculated s-SP and measured TSP values (in %).

<table>
<thead>
<tr>
<th>Struc.</th>
<th>Co</th>
<th>Fe</th>
<th>B</th>
<th>Avg. SP without B</th>
<th>Avg. SP with B</th>
<th>Exp. TSP</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-CoFeB</td>
<td>49.6</td>
<td>47.7</td>
<td>58.6</td>
<td>45.5</td>
<td>50.0 ± 0.2</td>
<td>53 ± 0.5</td>
</tr>
<tr>
<td>c-CoFeB</td>
<td>40.5</td>
<td>39.9</td>
<td>54.5</td>
<td>37.4</td>
<td>41.4 ± 0.5</td>
<td>44 ± 0.5</td>
</tr>
</tbody>
</table>

Figure 3(b) shows the total s-DOS of amorphous and fcc CoFeB, which confirms the higher SP of the amorphous alloy as given in Table I. If one compares the element-specific s-DOS for amorphous Co (and Fe—not shown) to fcc Co (and Fe) in Fig. 3(c), the antibonding s states of fcc Co (and Fe) are pushed towards higher energy for both spin channels. Increased s-d hybridization due to an increase in the first and second shell coordination of the fcc alloy might be responsible for this [Figs. 2(c) and 2(d)]. Interestingly, the decrease in the s-electron SP of the fcc alloy might be seen to primarily ensue from this spectral shift of the antibonding states toward higher energy, since $E_F$ lies on the slope of the increasing majority s-DOS, while lying in the deep minimum of the minority s-DOS. One notices from Fig. 3(b) that the minority DOS also shows subtle changes, which provide a secondary contribution to the change in the s-electron SP. The impact of s-d hybridization can also be seen in the B s-DOS shown in Fig. 3(d). In our calculations we note that (1) the B s p states are highly spin polarized (s-SP > 50%; p-SP > 25%) as noted before [22], and (2) the B sites attain a small negative magnetic moment (∼0.1µB) consistent with earlier work [23].

In summary, we show that in AlO$_x$ based junctions, the TSP of amorphous CoFeB is larger than that of fcc CoFeB. Calculations of the atomic and electronic structure of amorphous and fcc CoFeB yield s-electron SP values in remarkable agreement with experiment. These observations demonstrate that the electronic structure of the electrode has a marked impact on tunneling, and the SP of such a complex ternary amorphous-crystalline alloy can be genuinely calculated.

Research supported by NanoNed, FOM, STW via NWO-VICI grants, and ESTEEM—Contract No. 260019. We gladly acknowledge R. Coehoorn for fruitful discussions.

*p.v.paluskar@tue.nl

[22] R. Coehoorn (to be published).