Impact of interface crystallization on inelastic tunneling in Al/AlOx/CoFeB

Impact of interface crystallization on inelastic tunneling in Al/AlO$_x$/CoFeB

P. V. Paluskar,[a] F. L. Bloom, J. T. Kohlhepp, H. J. M. Swagten, and B. Koopmans
Department of Applied Physics, Center for NanoMaterials, and COBRA Research Institute, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

E. Snoeck
CEMES-CNRS, 29 rue Jeanne Marvig, B.P. 94347, F-31055 Toulouse Cedex 4, France

(Received 8 October 2007; accepted 2 November 2007; published online 26 November 2007)

We report the change in inelastic electron tunneling spectra (IETS) for Al/AlO$_x$/CoFeB/Al junctions when the structure of CoFeB at its interface with AlO$_x$ is intentionally changed from quasiamorphous to highly textured fcc. While for the quasiamorphous interface there are signs of the size quantization of magnons, the spectra for the fcc interface show distinct excitations at bias voltages associated with known surface magnon modes in fcc Co. These results demonstrate that IETS can be used as a tool to probe distinct structural changes of the magnetic electrode in tunnel junctions. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2817229]

Inelastic electron scattering processes, especially those involving magnetic excitations, have great influence on electronic transport in spintronic devices. In tunnel junctions, exchange scattering with localized impurities at the barrier interface are known to cause zero bias anomalies.[1] Applebaum's theory[2] on the origin of these anomalies has been applied to tunnel junctions,[3] and has been the key to explaining Kondo scattering at a single magnetic impurity.[4,5] Another important process considered by Appelbaum[2] which is highly relevant to magnetic tunnel junctions (MTJs) involves inelastic tunneling of hot electrons by exciting magnons at the barrier-electrode interface. Since such a spin-flip scattering process provides spin-mixing contributions to the total conductance, it is known to decrease tunnel magnetoresistance (TMR) in MTJs.[6,7]

Inelastic electron tunneling spectroscopy (IETS) is a powerful tool to isolate and identify excitation spectra from specific contributions to tunneling,[8,9] largely due to the fact that such processes have discrete threshold energies which result in peaks in d^2I/dV^2.[10] It has been used to observe that tunneling electrons excite phonons[10] and magnons[11] *in* the barrier. Indeed, it has also been employed by Moodera *et al.*[7] and by Nagahama *et al.*[12] to study tunneling in MTJs. Since exchange scattering with a magnon is directly related to the interfacial magnon modes, which in turn depend on the interface structure, IETS can be used to probe structural changes at the barrier-ferromagnet interface. No such observation has been reported as yet, presumably due to the difficulty of establishing such a marked structural change at the interface.

In this letter, we induce a distinct structural change in the ferromagnetic electrode at its interface with AlO$_x$, and thereafter probe the changes in the magnon spectrum using IETS. CoFeB is used as a ferromagnetic electrode primarily because the as-deposited quasiamorphous/nanocrystalline layer can be crystallized into a highly textured fcc layer by a single annealing on the same sample.[13] This allows for a straightforward comparison and a possibility of identifying specific contributions to inelastic tunneling. Moreover, CoFeB is of increasing importance, both fundamentally and technologically, as it contributes to huge TMR in AlO$_x$ (Ref. 14) and MgO (Ref. 15) based MTJs, and facilitates record-low switching currents in spin-torque devices.[16] From our IETS spectra, we notice that for amorphous electrodes, the small grain size at the interface might induce a low energy cutoff in the magnon spectrum due to size quantization effects. On crystallization in the fcc (111) texture, the increase in grain size lifts the size quantization and, we observe the appearance of a distinct peak in the spectra which might be directly related to known magnon excitations in single crystalline fcc Co.

We prepared Al/AlO$_x$/Co$_{72}$Fe$_{20}$B$_8$/Al junctions with CoFeB layer thickness (60 Å) specifically chosen to maximize their crystallization, particularly at the AlO$_x$ interface after a single annealing. One set of junctions from the same batch was annealed at T_a=450 °C in ultrahigh vacuum (pressure $<10^{-8}$ mbar during annealing). The IETS spectra were measured using a standard second harmonic lock-in technique at 4.2 K by applying a 3 mV/711 Hz ac modulation on top of a dc bias sweep (precision >100 μV).

To verify that CoFeB crystallized at the AlO$_x$ interface, we performed high-resolution transmission electron microscopy (HRTEM) on as-deposited and annealed samples. Figure 1(a) shows a junction in the as-deposited state, and (b) after an annealing at T_a=450 °C. For the as-deposited junction, a close inspection shows hardly any crystalline CoFeB at the AlO$_x$ interface. On the contrary, in the case of an

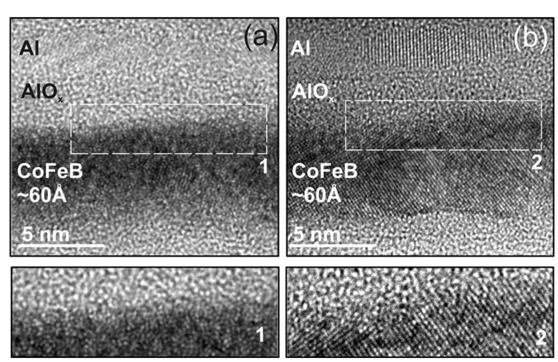

FIG. 1. HRTEM on Al/AlO$_x$/CoFeB (60 Å) junction before (a) and after (b) 450 °C annealing (see lower panels for zoom in).

[a]Electronic mail: p.v.paluskar@tue.nl

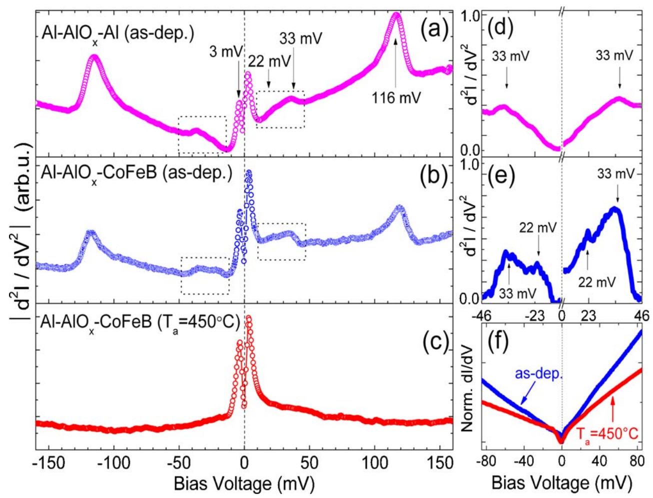

FIG. 2. (Color online) Representative IETS spectra for (a) as-deposited $Al/AlO_x/Al$ and (b) as-deposited $Al/AlO_x/CoFeB$. The dotted box indicates magnified regions in (d) and (e). Note the x-axis scale breaks. (c) shows spectra for the annealed junction. (f) The dI/dV for as-deposited and annealed CoFeB junction. The arrows are guides to the eye.

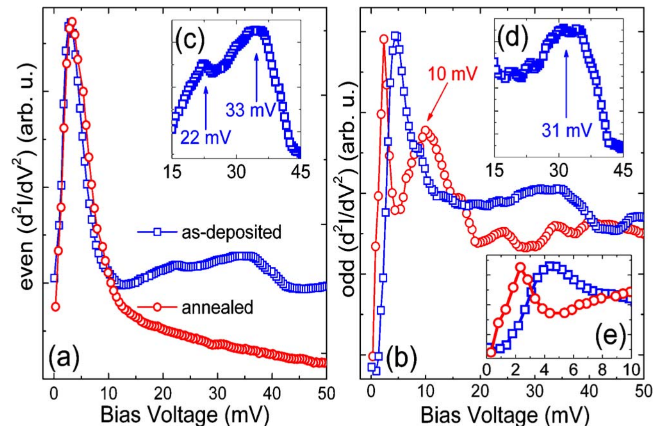

FIG. 3. (Color online) (a) Even and (b) odd spectra for an as-deposited (□) and annealed (○) $Al/AlO_x/CoFeB$ junction with insets (c) and (d) showing magnified Al phonon region. The inset (e) shows comparison of the odd zero bias anomaly region. The y-axis intensities are scaled to enable comparison.

annealed junction, we observe almost comprehensive crystallization of CoFeB in a fcc (111) texture, particularly at its interface with AlO_x. The bottom Al electrode is observed to be crystalline in both the as-deposited and annealed junctions.

Figure 2 shows representative IETS spectra for (a) an $Al/AlO_x/Al$ junction, (b) an as-deposited $Al/AlO_x/CoFeB/Al$ junction, and (c) a similar CoFeB junction annealed at $T_a=450$ °C. One notices that, in general, the intensity of d^2I/dV^2 and dI/dV [Fig. 2(f)] at positive bias, i.e., when electrons tunnel into the top electrode, is larger than that at a corresponding negative bias. This is generally attributed to barrier asymmetry.[17] The sharp peaks around ±3 mV in all three junctions (Figs. 2(a)–2(c), *follow the arrows in 2(a) as guides to the eye*) are generally assigned to the zero bias anomaly. The dI/dV [Fig. 2(f)] shows a sharp dip due to this anomaly. In the $Al/AlO_x/Al$ junction [Fig. 2(a)], apart from these sharp peaks, two sets of distinct shoulders can be seen: those around ±22 and ±33 mV correspond to Al TA and LA phonon modes, respectively,[18] and a sharp peak around ±116 mV corresponding to the OH bending mode of aluminum hydroxide.[18] Generically, the IETS spectra can be composed of contributions which are both symmetric and asymmetric with respect to the polarity of the bias voltage. This depends on the actual physical location of the excitations.[18] The presence of Al phonons around 22–33 mV at positive *and* negative bias indicates that their creation and annihilation by an electron tunneling into or out of the Al electrode has almost equal probability. Such symmetry under bias reversal has also been observed by Han *et al.*[19]

Although both sets of phonons, Al (22–33 mV) and OH (116 mV) are also seen in the as-deposited CoFeB junction [Fig. 2(b)], they are not observed in the annealed CoFeB junction [Fig. 2(c)]. The absence of the OH phonon *after* annealing has been noted before.[20] Presently, there is no insight in the absence of both these sets of peaks in the annealed junctions. However, from past experiments[13,21] one might conclude that their absence has no impact on the tunneling spin polarization of AlO_x based junctions. Also, one might wonder if the absence of the Al phonons (22–33 mV) in the annealed CoFeB junction [Fig. 2(c)] might be reflected in the superconducting properties of the Al electrode, presumably ensuing from structural or compositional changes in the films. However, we found no significant change in the superconducting gap, orbital depairing and spin-orbit scattering of superconducting Al electrodes at 0.27 K.

Turning to magnetic excitations, on a closer look at the Al phonon region for the $Al/AlO_x/Al$ junction [Fig. 2(d)], one does not observe any significant asymmetry in the intensity of the peaks under bias reversal. On the contrary, in the case of the *as-deposited* CoFeB junction [Fig. 2(e)], we find that the intensity of the peak at positive bias (+33 mV) is almost twice as large as that at negative bias (−33 mV). This asymmetry can be more clearly noticed if one looks at the odd and even d^2I/dV^2,[10]

$$\frac{d^2I}{dV^2}(\text{even/odd}) = \frac{d^2I}{dV^2}(+V) \pm \frac{d^2I}{dV^2}(-V). \qquad (1)$$

The even part of d^2I/dV^2 enhances the symmetric features, whereas these symmetric features cancel out in the odd part, leaving the asymmetric contributions with respect to bias polarity clearly portrayed. These are shown in Fig. 3. For the as-deposited CoFeB junction, while the peaks around 22 and 33 mV are readily identified in the even spectra [see square symbols in Figs. 3(c) and 3(a)], one would expect them to disappear in the odd spectra if only Al phonons were involved [see square symbols in Figs. 3(d) and 3(b)]. Instead, the odd spectra show a pronounced residual shoulder around 31 mV. This clearly suggests that in addition to Al phonon-assisted tunneling, for positive bias there is an added contribution to the tunnel conductance which has a threshold around +31 mV. One such possible contribution can be the onset of a sharp conduction band above the Fermi level of CoFeB which increases the phase space for the tunneling electrons at this bias. However, one does not expect such sharp changes in the electronic density of states (DOS) for a quasiamorphous ternary alloy.[13] An alternative explanation for this higher scattering intensity is magnon-assisted tunneling. One may anticipate that in a nanocrystalline material, as the grain size decreases, the coherence length of a magnon is increasingly limited[6] leading to size quantization and the appearance of a low energy cutoff in the magnon DOS.[22] Given that amorphous alloys follow the spin wave dispersion relation $\hbar\omega_k = Dk^2$,[23] a simple first order estimate of this low energy cutoff can be calculated as $E_{lc} = D(k_{x\,min}^2 + k_{y\,min}^2 + k_{z\,min}^2) \approx 3D(\pi^2/d^2)$ where $k = \pi/d$ and D is the exchange stiffness constant. For $E_{lc} = 31$ mV, and a grain size $d \approx 12-14$ Å calculated using the Scherrer formula on x-ray diffraction measurements, we obtain $D \approx 150-205$ meV Å^2. This value is in good agreement with 200 meV Å^2 measured for amorphous $Co_{80}B_{20}$ (Ref. 24) and 185 meV Å^2 for amorphous $Co_{80}P_{20}$.[23]

As the grain size is expected to increase after crystallization, one might expect suppression of the size quantization effect. Open circles in Fig. 3(b) show the odd spectra for the *annealed* CoFeB junction. Indeed, one notices that the peak around 31 mV is replaced by very small features at bias voltages above 20 mV, indicating that the quantization due to small grain sizes is lifted by the annealing. Remarkably, one also notices the appearance of a very distinct peak around 10 mV. Phonons of CoO,[19] Fe_3O_4,[25] and B_2O_3 (Ref. 26) have been measured at much higher energies (>45 meV). Thus, at this energy, one can rule out the formation of transition metal or boron oxide at the interface which leads to inelastic phonon-assisted tunneling. This argument is substantiated by the fact that we do not observe any significant postannealing change in junction resistance. Moreover, the tunneling spin polarization of these junctions with thick CoFeB films ($\gtrsim$500 Å) does not change after the annealing[13] and one does not find a strong argument as to why oxide formation should occur for thinner films. Furthermore, the presence of a sharp conduction band edge just above the Fermi level of fcc CoFeB (111) contributing to enhanced conductance in such a disordered ternary alloy is highly unlikely. Band structure calculations are concomitant with this argument.[13] We tentatively ascribe this peak to magnon excitations at the AlO_x–CoFeB interface. Such excitations have also been seen in single crystalline fcc Co (111) around a bias energy of 9–13 mV.[27,28] The strong similarities with the present results are endorsed by the fact that CoFeB crystallizes in highly textured (111) fcc structure. In agreement with Balashov *et al.*, the strong peak in the positive direction indicates that the magnon creation operator for an electron tunneling into the ferromagnet has a much larger expectation value than the corresponding coefficient for the magnon annihilation operator.

Parenthetically, we look the zero bias anomaly peak which appears around 2–4 mV in the odd spectra [see Fig. 3(e)]. As compared to the as-deposited CoFeB junction, this peak is much sharper and shifted to lower energies for the annealed CoFeB junction. This postannealing change in the peak might be due to the rearrangement of the localized magnetic impurity states in the barrier. One might wonder if the shift allows distinction between impurity assisted spin-flip tunneling[3] and magnon-assisted tunneling.[2] Experiments involving the dependence of the peak position on applied external magnetic fields at low temperatures might shed light on this issue.

In summary, we show that in Al/AlO_x/CoFeB based junctions, the IETS spectra are in sharp contrast depending on the structure of CoFeB at the interface. For amorphous CoFeB at the interface, we see indications of size quantization of the magnons. For fcc CoFeB at the interface, we see distinct excitations around 10 mV which could also be related to magnon-assisted spin flip tunneling. We demonstrate that the IETS technique is a powerful tool to investigate the impact of interface structure changes in magnetic tunnel junctions.

This work was supported by NanoNed, a Dutch nanotechnology program of the Ministry of Economic Affairs, and the HRTEM by IP3 project of the European Commission: ESTEEM Contract No. 0260019.

[1]A. F. G. Wyatt, Phys. Rev. Lett. **13**, 401 (1964).
[2]J. Appelbaum, Phys. Rev. B **183**, 553 (1972).
[3]L. Shen and J. Rowell, Phys. Rev. **165**, 566 (1968).
[4]V. Madhavan, W. Chen, T. Jamneala, M. F. Crommie, and N. S. Wingreen, Science **280**, 567 (1998).
[5]A. Heinrich, J. Gupta, C. Lutz, and D. Eigler, Science **306**, 466 (2004).
[6]S. Zhang, P. M. Levy, A. C. Marley, and S. S. P. Parkin, Phys. Rev. Lett. **79**, 3744 (1997).
[7]J. S. Moodera, J. Nowak, and R. J. M. van de Veerdonk, Phys. Rev. Lett. **80**, 2941 (1998).
[8]B. Stipe, M. Rezaei, and W. Ho, Science **280**, 1732 (1998).
[9]A. Heinrich, C. Lutz, J. Gupta, and D. Eigler, Science **298**, 1381 (2002).
[10]J. Lambe and R. Jaklevic, Phys. Rev. Lett. **17**, 1139 (1966).
[11]D. Tsui, R. Dietz, and L. Walker, Phys. Rev. Lett. **27**, 1729 (1969).
[12]T. Nagahama, S. Yuasa, Y. Suzuki, and E. Tamura, Appl. Phys. Lett. **79**, 4381 (2001).
[13]P. V. Paluskar (unpublished).
[14]H. X. Wei, Q. H. Qin, M. Ma, R. Sharif, and X. F. Han, J. Appl. Phys. **101**, 09B501 (2007).
[15]D. Djayaprawira, K. Tsunekawa, M. Nagai, H. Maehara, S. Yamagata, N. Watanabe, S. Yuasa, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **86**, 092502 (2005).
[16]J. Hayakawa, S. Ikeda, Y. M. Lee, R. Sasaki, T. Meguro, F. Matsukura, H. Takahashi, and H. Ohno, Jpn. J. Appl. Phys., Part 2 **44**, L1267 (2005).
[17]P. LeClair, J. T. Kohlhepp, H. J. M. Swagten, and W. J. M. de Jonge, Phys. Rev. Lett. **86**, 1066 (2001).
[18]E. L. Wolf, *Principles of Electron Tunneling Spectroscopy* (Oxford University Press, New York, 1989), 71, 219.
[19]X.-F. Han, J. Murai, Y. Ando, H. Kuboto, and T. Miyazaki, Appl. Phys. Lett. **78**, 2533 (2001).
[20]Y. Ando, J. Murai, H. Kuboto, and T. Miyazaki, J. Appl. Phys. **87**, 5209 (2000).
[21]C. H. Kant, J. T. Kohlhepp, H. J. M. Swagten, and W. J. M. de Jonge, Appl. Phys. Lett. **84**, 1141 (2004).
[22]K. Yakushiji, F. Ernult, H. Imamura, K. Yamane, S. Mitani, K. Takanashi, S. Takahashi, S. Maekawa, and H. Fujimori, Nat. Mater. **4**, 57 (2005), and Ref. 33 therein.
[23]H. Mook, N. Wakabayashi, and D. Pan, Phys. Rev. Lett. **34**, 1029 (1975).
[24]H. Watanabe, H. Morita, and H. Yamauchi, IEEE Trans. Magn. **14**, 944 (1978).
[25]L. V. Gasparov, D. B. Tanner, D. B. Romero, H. Berger, G. Margaritondo, and L. Forró, Phys. Rev. B **62**, 7939 (2000).
[26]Z. Wang, Y. Zhao, P. Lazor, H. Annersten, and S. K. Saxena, Appl. Phys. Lett. **86**, 041911 (2001), and Refs. 10–15 therein.
[27]T. Balashov, A. Takács, W. Wulfhekel, and J. Kirschner, Phys. Rev. Lett. **97**, 187201 (2006).
[28]R. Vollmer, M. Etzkorn, P. S. Anil Kumar, H. Ibach, and J. Kirschner, Phys. Rev. Lett. **91**, 147201 (2003).