Journal of Magnetism and Magnetic Materials 242–245 (2002) 47–52

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

Tunneling magnetoresistance through a vacuum gap

W. Wulfhekel*, H.F. Ding, J. Kirschner

Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany

Abstract

We studied the tunneling magnetoresistance (TMR) effect through a vacuum barrier using spin-polarized scanning tunneling microscopy on Co(0 0 0 1). By varying the gap width at a fixed bias voltage or by varying the bias voltage at a fixed gap width, the fundamental behaviour of the TMR across the vacuum gap was investigated. At large gap widths the TMR is constant with the width in agreement with Jullières model. At gap widths below ≈ 4.5 Å, a decrease of the TMR was found which cannot be explained on the basis of this model. The decrease is correlated with a strong decrease of the local barrier height underneath the tip and is explained in the framework of Slonczewski's model. The TMR across the vacuum barrier does not show the characteristic drop with bias voltage usually found in planar tunneling junctions but is rather independent on the voltage in case of large gap widths. This is related to the tunneling of electrons predominantly perpendicular to the Co(0 0 0 1) surface and the particular band structure of Co. The voltage dependence, however, is more complex at small gap width due to the opening up of the emission cone of the tunneling electrons.

Keywords: Tunneling; Magnetoresistance; Spin polarization; Band structure—energy

In 1975, Jullière found that the tunneling resistance between two ferromagnetic layers separated by an insulating barrier, depends not only on the bias voltage but also on the relative orientation of magnetization of the two magnetic layers [1]. The tunneling magnetoresitance (TMR) effect was discovered. The recent success of reproducible fabrication of high-quality tunneling junctions operating at ambient temperatures [2], has prompted intensive studies on the underlying physical mechanisms [3–8] and has stimulated many applications in magnetic sensors or magnetic random access memory [9–12]. In spite of the intensive fundamental research and the promising applications, the TMR effect is still not completely understood and many open questions need to be addressed. One difficulty to obtain a coherent picture of the effect is related to the complex structure of the tunneling junctions, often containing poorly characterized amorphous barriers causing higher order effects in spin-polarized tunneling [10,13,14]. Here, we study the TMR effect across a simple barrier, the vacuum gap of a spin-polarized scanning tunneling microscope (Sp-STM) [15]. Sp-STM provides a model system to test different theories for spin-polarized tunneling as it has a simple vacuum barrier, whose width can be controlled in a precise way, as well as it allows to use atomically clean surfaces as electrodes. With the use of Sp-STM, we would like to answer two open questions. Firstly, where are the limits of Jullière's model, and secondly, is the generally observed strong voltage dependence of the TMR effect of planar tunneling junctions [1,2,16] also found for the ideal vacuum barrier of the Sp-STM?

2. Experimental setup

A bulk single crystalline Co(0 0 0 1) sample was chosen as one of the electrodes. The second electrode was a magnetic tip of the scanning tunneling microscope. The material of the tip is an amorphous Co based alloy [17] with extremely low coercive fields (<1 Oe) and vanishing magnetostriction ($<10^{-8}$). During scanning the tip over the surface of the Co crystal, an alternating current of 40 kHz was passed through a small coil wound around the magnetic tip to periodically switch

*Corresponding author. Tel.: +49-345-55-82-674; fax: +49-345-55-11-223.

E-mail address: wulf@mpi-halle.mpg.de (W. Wulfhekel).

0304-8853/02/$ - see front matter
PII: S 0 3 0 4 - 8 8 5 3 (0 1) 0 1 1 8 2 - 9

the longitudinal magnetization of the tip. This results in modulations of the tunnel current due to the TMR effect [1]. These modulations were detected with a phase sensitive lock-in amplifier to map the magnetic structure of the sample [15,18,19]. The vanishing magnetostriction of the material ensures that no significant vibrations of the tip occur [19,20]. In this way, the variation of the tunneling current is measured, while the feed back loop keeps the average tunneling current constant, i.e., the current asymmetry δ caused by the TMR effect (see below) can be directly obtained by dividing the lock-in signal by the mean tunneling current of the STM. The detailed principle and technique of Sp-STM have been addressed in previous publications [15,18,19]. All experiments were performed in ultra-high vacuum ($p = 5 \times 10^{-11}$ mbar). The vacuum chamber was equipped with an Auger electron spectrometer (AES), low-energy electron diffraction (LEED) and a Sp-STM [15,18,19]. Both the Co(0 0 0 1) sample and the magnetic tip were cleaned in situ by sputtering with 1 keV Ar^+ ions. The sample was annealed afterwards to 570 K. In AES spectra of the sample no traces of contaminations could be found. LEED images showed the expected sixfold diffraction pattern with sharp spots and low background intensity. Tunneling images of both the topography and the magnetic structure were recorded simultaneously at room temperature. The typical dendritic like perpendicular domain pattern of Co(0 0 0 1) was observed, similar to that seen with magnetic force microscopy [21] and scanning electron microscopy with polarization analysis [22]. To study the TMR as a function of the barrier width or gap voltage, we zoomed into a small area which contains only two domains and one domain wall and imaged the same scan line repeatedly as a function of the vacuum barrier width or the gap voltage. For the measurements of the gap width dependence, in each pixel of the scan line the feedback loop was opened for a short time and the tip was approached/retracted continuously up to a displacement of 0.1 nm from its original position while measuring the average tunneling current and the modulated current caused by the TMR effect. The current asymmetry δ is calculated from their ratio. For the measurements of the voltage dependence, we proceeded in a similar way. The feedback loop was opened and the sample bias was ramped while recording the average tunneling current and the modulated current to obtain δ.

3. The limits of Jullière's model

Although there are many models to explain the TMR effect, there is wide consensus that it is a consequence of the exchange splitting of the band structure of the ferromagnetic electrodes which leads to an unbalanced distribution of majority and minority electrons at the Fermi energy. Assuming the conservation of the electron spin during tunneling this imbalance leads to different conductance G for parallel (p) and antiparallel (a) magnetic configuration. Jullière [1] proposed a phenomenological model that relates the TMR, or as we would like to phrase it here the conduction asymmetry δ, to the spin-polarization P_i at the Fermi energy of the two electrodes. Under the assumption of low bias voltage across the junction δ is given by

$$\delta = \frac{G_p - G_a}{G_p + G_a} = P_1 P_2. \tag{1}$$

This model has been commonly used in many studies [2,3], sometimes with the extension that the polarization P_i is not the spin-polarization of the ferromagnet but that of the ferromagnet/barrier interface [5–8]. To test this model, we study the dependence of the TMR on the gap width. In Jullière's model, there is no implicit or explicit dependence on the barrier properties. The TMR is entirely determined by the density of states of the electrodes. Fig. 1 shows four typical magnetic images taken of a domain wall on Co(0 0 0 1) and the corresponding line profiles (δ as a function of position) for the different tip displacements. Zero displacement corresponds to the gap distance at 20 mV gap voltage and 5 nA tunneling current ($\approx$4.5 Å gap width, see below) and negative displacements correspond to an approach of the tip towards the surface. From the images, it is obvious that the TMR is a function of the tip-to-sample distance in contrast to the prediction of Jullière. At small vacuum gap widths, the TMR almost vanishes. In the framework of Jullière's model, this corresponded to a vanishing of one or both spin-polarizations of the electrodes, i.e., at short distances they became non magnetic. This of course is not the case. Instead, it seems that Jullière's model fails here.

To explain this unexpected drop of the TMR with gap width, we have a closer look at the distance dependence of both the tunneling current and the current asymmetry δ. Fig. 2a shows a typical tunneling current versus the tip displacement obtained at 20 mV sample bias. The tunneling current increases nearly exponentially when the tip approaches the sample surface as expected. Fig. 2b presents the current asymmetry δ as a function of the tip displacement. It shows that the TMR is nearly constant at large tip-to-sample distances and decreases at shorter distances when the tunneling resistance is smaller than 5 MΩ (20 mV, 4 nA). Assuming a contact resistance of $\approx$24 kΩ, the resistance of 5 MΩ corresponds to a tip-to-sample distance of $\approx$4.5 Å [23–26]. As all other tunneling parameters are kept constant during the measurement, the variation of the TMR has to be attributed to the change of the tip-to-sample distance, either directly or indirectly via changes of other barrier properties induced by it.

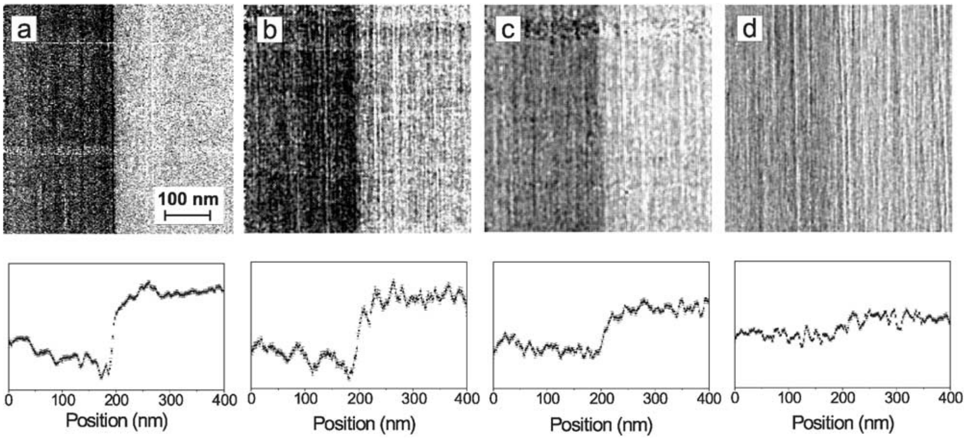

Fig. 1. Magnetic images and line scans of the current asymmetry δ across a domain wall on Co(0 0 0 1) obtained by Sp-STM with different tip displacements. (a) +1 Å, (b) +0.5 Å, (c) 0 Å, (d) −0.5 Å. The bias voltage is 20 mV in all images. Zero displacement corresponds to the gap distance of ≈4.5 Å.

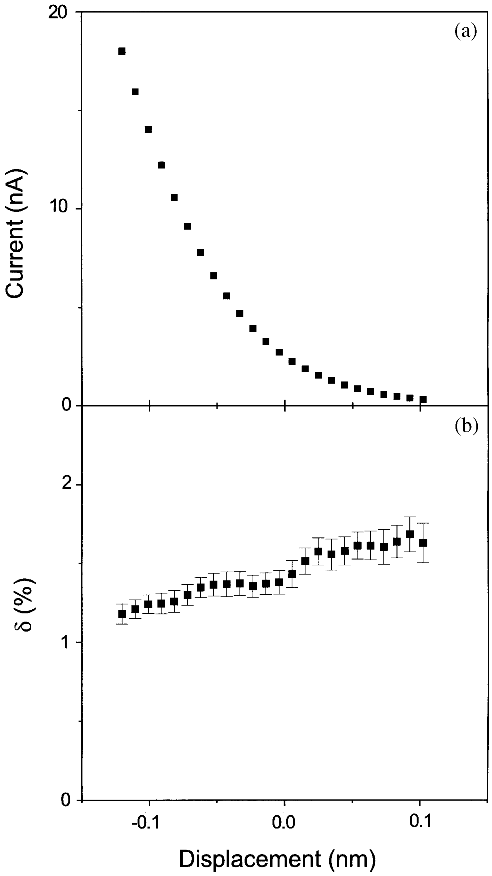

Fig. 2. (a) Tunneling current as a function of tip displacement. Negative displacements indicate an approach of the tip from the original position corresponding to tunneling parameters of 5 nA at 20 mV. (b) Simultaneously measured current asymmetry δ.

In the free electron approximation, Slonczewski calculated the TMR and pointed out that it does not only depend on the two ferromagnetic electrodes but also on the barrier [27]. Instead of using the spin-polarizations of the electrodes, he proposed to use an effective spin polarization of the ferromagnetic electrode and barrier couple P_{fb} to describe the TMR effect:

$$\delta = P_{1\mathrm{b}} P_{2\mathrm{b}}, \tag{2}$$

where $P_{1\mathrm{b}}$ and $P_{2\mathrm{b}}$ are defined as

$$P_{1(2)\mathrm{b}} = \frac{(k^{\uparrow}_{1(2)} - k^{\downarrow}_{1(2)})}{(k^{\uparrow}_{1(2)} + k^{\downarrow}_{1(2)})} \frac{(\kappa^2 - k^{\uparrow}_{1(2)} k^{\downarrow}_{1(2)})}{(\kappa^2 + k^{\uparrow}_{1(2)} k^{\downarrow}_{1(2)})} \tag{3}$$

The first factor in this equation represents the spin-polarization of the electrodes at the Fermi energy given by the wave vectors inside the ferromagnetic electrodes $k^{\uparrow}_{\mathrm{f}}$ and $k^{\downarrow}_{\mathrm{f}}$ of majority and minority electrons. The second factor represents a correction factor to the simple spin-polarization depending on $i\kappa$, the imaginary wave vector of the tunneling electrons in the barrier. In the limit of small bias voltage, it is defined by $\hbar\kappa = [2m(V_{\mathrm{b}} - E_{\mathrm{F}})]^{1/2}$, ($V_{\mathrm{b}}$ is the barrier height). Hence, through κ, the TMR depends on the height of the barrier V_{b}. When the local barrier height changes with the tip-to-sample distance, the TMR also changes. It is well known that the local barrier height in STM measurements decreases when the tip is approached closer than ≈4 Å [23–26]. The decrease is due to the fact that the electron densities of the tip and the sample start to overlap significantly and the tunneling electrons do not have to overcome the full work function but only a fraction of it. At small bias voltages, the local barrier height ϕ can be obtained from the tunneling current I as a function of the tip-to-sample distance according to the following equation [26]:

$$\phi(\mathrm{eV}) = 0.952\left(\frac{\mathrm{d}\ln \mathrm{I}}{\mathrm{d}S}\right)^2, \tag{4}$$

where the barrier width S is in Å. Fig. 3a presents the local barrier height versus the tip displacement calculated from the data shown in Fig. 2a. It is nearly constant at large tip-sample separations and decreases when the tip further approaches the sample. The observed change of local barrier height behaves similarly to the tip-to-sample distance-dependent TMR effect shown in Fig. 2b. This suggests a correlation between the barrier height and the TMR effect.

To quantify the influence of the local barrier height on the TMR effect, we performed calculations in the free electron model proposed by Slonczewski [27]. With the local barrier height measured above, the imaginary wave vector inside barrier for electrons tunneling near the Fermi level is determined. Therefore, applying the formula given in Eqs. (2) and (3), the TMR effect as a function of the tip displacement is calculated. As one parameter, the exchange energy is chosen to be 1 eV for Co [28]. Since there is no direct measurement for the spin-polarization of single crystal Co(0 0 0 1), we calculate the TMR for three different values, that are: 33% chosen from early measurement by Meservey and Tedrow [29], 45% chosen from recently reported values by Moodera et al. [16], and 66% for comparison reasons. The same values are chosen for the magnetic tip as the tip material is dominated by Co. The values mentioned above were obtained with Al_2O_3 barriers with a barrier height of ≈ 2.5 eV [16]. Using Eq. (3) and the exchange energy of 1 eV the wave vectors for spin up and spin down electrons in Co were calculated. With these three different sets of wave vectors and the distance dependent local barrier height, δ as a function of the tip-to-sample distance is calculated. Fig. 3b presents the results of the calculations. For comparison, the experimentally measured tip-to-sample distance-dependent current asymmetries (filled squares) are shown in this figure as well. All curves are normalized to the asymmetry value at large tip-to-sample distance. The figure shows that the calculations for all three spin-polarizations reproduce well the decrease of the TMR with the tip approaching, even though the polarizations are varying by a factor of 2. Hence, the observed distance dependence of the TMR can be explained well on the basis of Slonczewski's model, where the decrease of the TMR is not related to a change of the spin-polarization of the electrodes but to the change of the barrier properties at small gap widths. We believe that in general, the dependence of the barrier height should be included in the discussion of the TMR also in planar tunneling junctions. The mere density of states of the electrodes in Jullière's model is not sufficient to fully explain the size of the TMR effect.

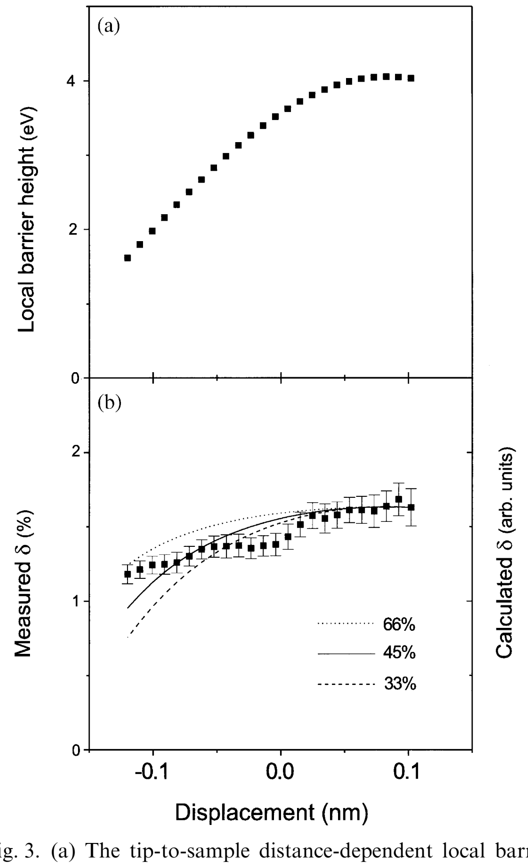

Fig. 3. (a) The tip-to-sample distance-dependent local barrier height calculated from the tunneling current shown in Fig. 2a. (b) Comparison between the measured current asymmetry δ and the calculated using the free electron model proposed by Slonczewski with the local barrier height given in (a) for three different values of the spin polarization. The calculated curves are normalized to the measured δ at large tip-to-sample distance.

4. Bias voltage dependence of the TMR

As the second fundamental aspect of the TMR effect, we would like to address its voltage dependence. In planar tunneling junctions, mostly using amorphous or polycrystalline barrier materials like Al_2O_3 [16], a rather strong drop of the TMR is observed with increasing the bias voltage [1,2,16]. For junctions of similar electrodes but different insulators or even same insulators but prepared in different ways, very different voltage dependences have been observed. In the early work of Jullière, only 3 mV bias was needed to halve the TMR of the junction [1]. Later, Moodera et al. fabricated junctions, were 200 mV were needed [2]. With increased control of the barriers, this value has been increased considerably to 500 mV [8] or even 700 mV [10]. Many different models have been proposed that explain this drop of the TMR with bias voltage. Biasing the junctions leads to tunneling from electrons mostly around the Fermi energy of the negative electrode to empty states at energies of the bias voltage above the Fermi energy in the positive electrode. The spin-polarization of the empty states, however, does not need to be constant and variations of it should cause variations of the TMR. This so called density of states effect has been observed in crystalline junctions [5]. Secondly, the hot electrons in the positive electrode might be spin-scattered by interfacial spins [4] or might create magnons [30], both leading to the loss of spin-information and reduction of the TMR. Finally, as Zhang and White suggested, the insulator barriers might contain localized electronic defects such that tunneling through the barrier does not proceed in a coherent way but the electrons tunnel via trap states reducing the spin-polarization [13]. This model is also supported by others based on experiments or theory [8,31,32]. In the case of a vacuum barrier, this mechanism should not be operating, however, magnon creation, spin scattering at the interface and the density of states effect in principle could be present. Fig. 4a shows the current asymmetry δ as a function of the gap voltage for large tip-to-sample distance (feed back parameters 1 nA, 1V). Obviously, the TMR is not a strong function of the bias voltage and only slightly decreases with raising the bias to ± 0.9 V. This is in sharp contrast to the drop seen in planar tunneling junctions. For tunneling into Co(0 0 0 1), the role of spin scattering at the interface and magnon creation in the bulk seems of no major importance. They do not cause a strong drop in our case. Also the spin-polarization of the density of states does not seem to vary much even with rather high bias voltages. This, however, is expected from the theoretical point of view. For large gap widths, mostly electrons with a perpendicular moment to the surface contribute to the tunneling current [27,33–35] and the emission cone of the STM is rather focused. Therefore, when tunneling into the empty states of the Co(0 0 0 1) electrode, mostly those states are sampled that are along the ΓA direction. Along this direction, however, there are only two minority bands and no majority states available in Co(0 0 0 1) [36,37]. The spin-polarization, hence, should be rather constant and the TMR should not vary in agreement with our experiment. For opposite bias, the electrons tunnel into the amorphous tip which should not display sharp features in its empty density of states. The rather constant TMR is not found for all gap widths. For small gap widths, the emission cone of STM opens up and also electrons with non normal momentum contribute significantly to the tunneling current [27,33–35]. In that case, a different voltage dependence can be observed in the current asymmetry δ. The TMR depends strongly on the voltage (see Fig. 4b taken with feed back parameters of 1 nA, 100 mV). There is a dip of the TMR around 200 mV bias. Possibly, this dip is related to majority states which lie off normal. Due to their majority nature, the spin-polarization is reduced and the TMR is reduced as well. We are currently performing first principle calculations to pinpoint the exact states that contribute to the tunneling current [38].

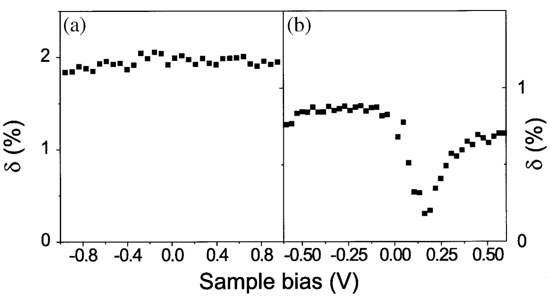

Fig. 4. Voltage dependence of the current asymmetry δ for large (a) and small (b) vacuum gap width.

From the voltage dependence of the TMR one can learn that it is not the density of states that dominate the size of the TMR but one has to be more careful. One has to consider the full band structure to explain the voltage dependence and the direction of the tunneling electrons have to be considered as well. This has also been pointed out from theory, recently [33–35,39]. The origin of the rather strong drop of the TMR in planar tunneling junctions still stays on open question. However, on the basis of our findings, one might conclude that most of the voltage dependence is related to the elctronic structure of the amorphous barrier containing localized trap states [13] and not related to magnon creation or spin excitations at the interface.

5. Conclusions

In conclusion, using a model system with a tunable vacuum barrier we measured both the gap width and voltage-dependent TMR, we show the limits of Jullière's model and illuminate the importance of details of the band structure for the TMR effect. For the dependence of the TMR on the gap width, Slonczewski's model is found to be a good description. For the voltage dependence, the details of the band structure and not only the density of states are needed to get a full understanding. We hope that with these measurements more theoretical and experimental work on the TMR effect is motivated leading to a better understanding of the fundamental processes during spin-polarized tunneling in Sp-STM and also planar tunneling junctions.

Acknowledgements

The authors acknowledge discussions with J. Henk, P. Bruno and I. Mertig.

References

[1] M. Jullière, Phys. Lett. 54 A (1975) 225.
[2] J.S. Moodera, L.R. Kinder, T.M. Wong, R. Meservey, Phys. Rev. Lett. 74 (1995) 3273.
[3] Y. Lu, R.A. Altman, A. Marley, S.A. Rishton, P.L. Trouilloud, G. Xiao, W.J. Gallagher, S.S.P. Parkin, Appl. Phys. Lett. 70 (1997) 2610.
[4] S. Zhang, P.M. Levy, A.C. Marley, S.S.P. Parkin, Phys. Rev. Lett. 79 (1997) 3744.
[5] J.M.D. Teresa, A. Barthélémy, A. Fert, J. Contour, F. Montaigne, P. Seneor, Science 286 (1999) 507.
[6] M. Sharma, S.X. Wang, J.H. Nickel, Phys. Rev. Lett. 82 (1999) 616.
[7] P. LeClair, J.T. Kohlhepp, H.J.M. Swagten, W.J.M. de Jonge, Phys. Rev. Lett. 86 (2001) 1066.
[8] S. Yuasa, T. Sato, E. Tamura, Y. Suzuki, H. Yamamori, K. Ando, T. Katayama, Europhys. Lett. 52 (2000) 344.
[9] M. Tondra, J.M. Daughton, C. Nordman, D. Wang, J. Taylor, J. Appl. Phys. 87 (2000) 4679.
[10] H. Boeve, E. Girgis, J. Schelten, J.D. Boeck, G. Borghs, Appl. Phys. Lett. 76 (2000) 1048.
[11] R. Scheuerlein, W. Gallagher, S.S.P. Parkin, A. Lee, S. Ray, R. Robertazzi, W. Reohr, IEEE International Solid-State Circuits Conference, Digest of Technical Papers, 2000.
[12] P.P. Freitas, S. Cardoso, R. Sousa, K. Wanjun, R. Ferreira, V. Chu, J.P. Conde, IEEE Transactions on Magnetics 36 (2000) 2796.
[13] J. Zhang, R. White, J. Appl. Phys. 83 (1998) 6512.
[14] E.Y. Tsymbal, D. Pettifor, J. Appl. Phys. 85 (1999) 5801.
[15] W. Wulfhekel, J. Kirschner, Appl. Phys. Lett. 75 (1999) 1944.
[16] J.S. Moodera, G. Mathon, J. Magn. Magn. Mater. 200 (1999) 248.
[17] M. Vázquez, J. González, A. Hernando, J. Magn. Magn. Mater. 53 (1986) 323.
[18] W. Wulfhekel, H.F. Ding, J. Kirschner, J. Appl. Phys. 87 (2000) 6475.
[19] W. Wulfhekel, H.F. Ding, W. Lutzke, G. Steierl, M. Vázquez, P. Marín, A. Hernando, J. Kirschner, Appl. Phys. A 72 (2001) 463.
[20] W. Wulfhekel, R. Hertel, H.F. Ding, J. Kirschner, J. Magn. Magn. Mater., submitted for publication.
[21] H.F. Ding, W. Wulfhekel, C. Chen, J. Barthel, J. Kirschner, Materials Science and Engineering B 84 (2001) 96.
[22] J. Unguris, M.R. Scheinfein, R.C. Celotta, D. Pierce, Appl. Phys. Lett. 55 (1989) 2553.
[23] J.K. Gimzewski, R. Moller, Phys. Rev. B 36 (1987) 1284.
[24] N.D. Lang, Phys. Rev. B 36 (1987) 8173.
[25] N.D. Lang, Phys. Rev. B 37 (1988) 10 395.
[26] Y. Kuk, P.J. Silverman, J. Vac. Sci. Technol. A 8 (1990) 289.
[27] J.C. Slonczewski, Phys. Rev. B 39 (1989) 6995.
[28] M. Stearns, J. Magn. Magn. Mater. 5 (1977) 167.
[29] R. Meservey, P.M. Tedrow, Solid State Commun. 11 (1972) 333.
[30] J.S. Moodera, J. Nowak, R.J.M. van de Veerdonk, Phys. Rev. Lett. 80 (1997) 2941.
[31] R. Jansen, J.S. Moodera, Phys. Rev. B 61 (2000) 9047.
[32] E.Y. Tsymbal, D.G. Pettifor, Phys. Rev. B 58 (1998) 432.
[33] J.M. MacLaren, X.G. Zhang, H.W. Butler, X. Wang, Phys. Rev. B 59 (1999) 5470.
[34] W.H. Butler, X.G. Zhang, T.C. Schulthess, J.M. MacLaren, Phys. Rev. B 63 (2001) 054416.
[35] J. Mathon, A. Umerski, Phys. Rev. B 63 (2001) 220403.
[36] T. Jarlborg, M. Peter, J. Magn. Magn. Mater. 42 (1984) 89.
[37] M. Getzlaff, J. Bansmann, J. Braun, G. Schönhense, J. Magn. Magn. Mater. 161 (1996) 70.
[38] H.F. Ding, W. Wulfhekel, J. Henk. P. Bruno, J. Kirschner, to be published.
[39] Ph. Mavropoulos, N. Papanikolaou, P.H. Dederichs, Phys. Rev. Lett. 85 (2000) 1088.