

Nanosized corners for trapping and detecting magnetic nanoparticles

To cite this article: Marco Donolato *et al* 2009 *Nanotechnology* **20** 385501

View the [article online](#) for updates and enhancements.

Related content

- [Detection of a single synthetic antiferromagnetic nanoparticle with an AMR nanostructure: Comparison between simulations and experiments](#)
M Donolato, M Gobbi, P Vavassori *et al*.
- [Synthesis and applications of magnetic nanoparticles for biorecognition and point of care medical diagnostics](#)
Adarsh Sandhu, Hiroshi Handa and Masanori Abe
- [Magnetoresistive sensors](#)
P P Freitas, R Ferreira, S Cardoso *et al*.

Recent citations

- [Magnetophoretic induced convective capture of highly diffusive superparamagnetic nanoparticles](#)
M. Fratzl *et al*
- [Magnetic Biosensors: Modelling and Simulation](#)
Vahid Nabaei *et al*
- [Controllable synthesis of anisotropic silica/polymer composite particles via seeded dispersion polymerization](#)
Zhen Zhang *et al*

Nanosized corners for trapping and detecting magnetic nanoparticles

Marco Donolato¹, Marco Gobbi¹, Paolo Vavassori^{2,3},
Marco Leone¹, Matteo Cantoni¹, Vitali Metlushko⁴, Bojan Ilic⁵,
Mingliang Zhang^{6,7}, Shan X Wang^{6,7} and Riccardo Bertacco¹

¹ LNESS—Dipartimento di Fisica, Politecnico di Milano, Via Anzani 42, I-22100 Como, Italy

² CIC nanoGUNE Consolider, E-20009 San Sebastian, Spain

³ CNR-INFN National Research Center S3, CNISM and Dipartimento di Fisica,
Università di Ferrara, I-44100 Ferrara, Italy

⁴ Department of Electrical and Computer Engineering, University of Illinois at Chicago,
Chicago, IL 60607, USA

⁵ Cornell Nanofabrication Facility, Cornell University, Ithaca, NY 14853, USA

⁶ Department of Materials Science and Engineering, Stanford University, Stanford,
CA 94305, USA

⁷ Department of Electrical Engineering, Stanford University, Stanford, CA 94305, USA

E-mail: marco.donolato@gmail.com

Received 9 April 2009, in final form 5 August 2009

Published 28 August 2009

Online at stacks.iop.org/Nano/20/385501

Abstract

We present a device concept based on controlled micromagnetic configurations in a corner-shaped permalloy nanostructure terminated with two circular disks, specifically designed for the capture and detection of a small number of magnetic beads in suspension. A transverse head-to-head domain wall (TDW) placed at the corner of the structure plays the role of an attracting pole for magnetic beads. The TDW is annihilated in the terminating disks by applying an appropriate magnetic field, whose value is affected by the presence of beads chemically bound to the surface. In the case where the beads are not chemically bound to the surface, the annihilation of the TDW causes their release into the suspension. The variation of the voltage drop across the corner, due to the anisotropic magnetoresistance (AMR) while sweeping the magnetic field, is used to detect the presence of a chemically bound bead. The device response has been characterized by using both synthetic antiferromagnetic nanoparticles (disks of 70 nm diameter and 20 nm height) and magnetic nanobeads, for different thicknesses of the protective capping layer. We demonstrate the detection down to a single nanoparticle, therefore the device holds the potential for the localization and detection of small numbers of molecules immobilized on the particle's functionalized surface.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In recent years magnetic beads and magnetic nanoparticles have received great interest for their applications in biology and nanomedicine [1]. For example, magnetic nanoparticles with functionalized surfaces are used as labels for single molecule study [2], cell manipulation [3] and biomagnetic sensing [4]. In the latter case, magnetic beads are employed to detect target molecules at the surface of magnetic sensors where probe molecules are immobilized.

For this particular application, various very sensitive magnetoresistive (MR) sensors have been proposed, based on giant magnetoresistance GMR [5], tunnel magnetoresistance TMR [6], miniaturized silicon Hall sensors [7] or the planar Hall effect in permalloy thin films [8].

The integration of the MR sensor in miniaturized platforms for collecting, manipulating and detecting magnetic nanoparticles has also been widely investigated as a potential pathway to lab-on-chip technology. Along this line, microelectromagnet arrays [9] and current lines [10] have been

used for the trapping and controlled movement of magnetic particles, and an integrated microfluidic cell for manipulating and detecting magnetic beads has been proposed by Jiang *et al* [11], using an anisotropic magnetoresistance (AMR) ring sensor as a microbead counter. In none of these integrated systems has the detection of a single magnetic nanoparticle been shown. On the other hand the detection of a single magnetic nanoparticle has been demonstrated employing InAs quantum well Hall devices [12] and magnetic tunnelling junctions [13], but without any focusing action of the particles on the sensor.

Recently, we have demonstrated the possibility of detecting a single 80 nm bead by measuring the AMR signal related to the displacement of a domain wall between two corners of a permalloy micron-sized square ring [14] initially designed for data-storage applications.

In this work we present an original magnetic particles nanosensor based on the principle described in [14], but specifically designed to present a single switchable magnetic pole for trapping magnetic beads on the detection area only. The combination of single magnetic nanoparticle detection and attraction on the active sensor area represents a peculiar feature in comparison to other sensing techniques. The switchable magnetic pole is implemented by a transverse domain wall (TDW) on the single corner of the structure, which is nucleated and annihilated via the application of magnetic fields. The particle attraction and the sensing properties of the device have been characterized by using both commercial beads and synthetic antiferromagnetic nanoparticles (SAF), for different thicknesses of a SiO₂ protective capping layer, with a view to applications in liquid environments.

2. Trapping of magnetic beads at the corner

The device consists of a patterned L-shaped permalloy strip, 25 nm thick and 180 nm wide, with two end disks (diameter 500 nm) that have their centre at 2 μ m from the corner (see figure 1(a)).

This nanostructure was fabricated on top of four 20 nm thick and 100 nm wide Au contacts (labelled 1–4 in figure 1(a)) on a Si/SiO₂ substrate. Figures 1(b) and (c) show the two stable micromagnetic configurations employed here, as obtained by using the object oriented micromagnetic framework (OOMMF) code [15].

The four leads (1–4) have been used to measure the voltage drop across the corner, which reveals the presence of a TDW thanks to the anisotropic magnetoresistance effect. In fact, when a TDW is in the corner, the current density $\mathbf{J}$ and the magnetization $\mathbf{M}$ are locally perpendicular (micromagnetic configuration in figure 1(b)) and the resistivity between leads 2 and 3 is lower than when the TDW is absent (micromagnetic configuration in figure 1(c)).

Figure 1(b) illustrates the creation of a TDW after the application of an external field of 1000 Oe along the bisector of the corner. In equilibrium a vortex configuration is present at the two end disks, corresponding to a clockwise and anticlockwise rotation of the magnetization. The TDW can be pushed towards one of the termination disks, where

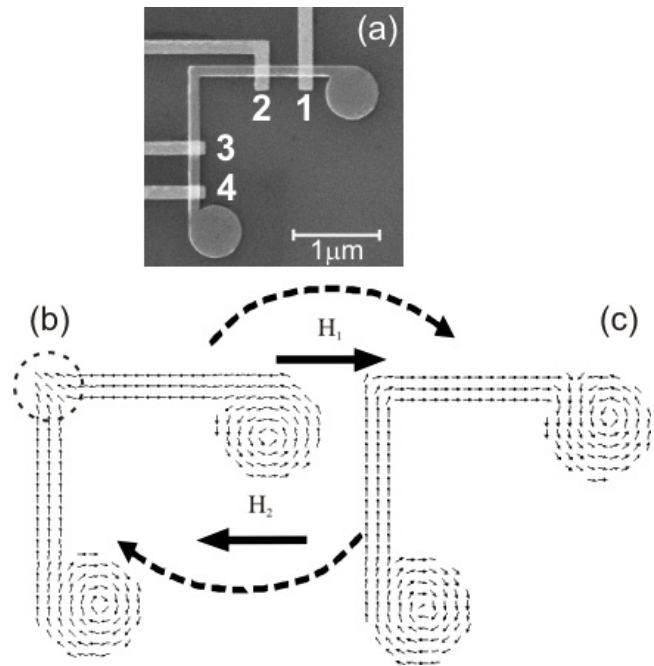


Figure 1. (a) SEM image of the device structure with the four leads to perform MR measurements. Micromagnetic configuration of the device (calculated with OOMMF) with (b) and without (c) TDW. H_1 and H_2 represent the fields required to switch from (b) to (c) and back, respectively.

it annihilates in the vortex spin configuration (simply VS hereafter) inside one of the two termination disks. This occurs on applying a magnetic field H_1 directed as indicated in figure 1, leading to the spin structure of figure 1(c). The TDW can be repositioned at the corner, either by applying a horizontal magnetic field H_2 in the opposite direction with respect to H_1 , or by again applying a field along the bisector of the corner.

Figures 2(a) and (b) show the contour plots of the modulus of the attractive force acting on a point magnetic moment $\mu(H)$ on a plane 200 nm from the sensor. The force originates from the gradient of the magnetic stray field $\mathbf{H}$ generated by the TDW, which results in an attractive force on any magnetic nanoparticle that is moving in proximity to the TDW location. In figure 2(a) the TDW is present at the corner, while in figure 2(b) it has been annihilated in the vortex spin configuration. These plots were produced by using the 3D OOMMF's calculated external field and the force expression $\mathbf{F} = \mu_0(\mu \cdot \nabla)\mathbf{H}$; here $\mu = \mu(H)\mathbf{h}$, where $\mu(H)$ is the magnetization curve of the bead and $\mathbf{h}$ is the unit vector parallel to the field $\mathbf{H}$. The function $\mu(H)$ was assumed to be a Langevin curve suitable for describing the behaviour of 130 nm MICROMOD Nanomag[®]-D beads ($M_s = 200 \text{ emu cm}^{-3}$).

The plots show that, in the presence of the TDW, such a bead is subjected to a force higher than 20 pN within a circular area of about 200 nm in diameter centred on the external vertex of the corner and placed 200 nm above the permalloy surface. The simulations show that there is also an attractive force localized in correspondence to the strip-disk connection, which is however much less intense (about 10% of that on the corner).

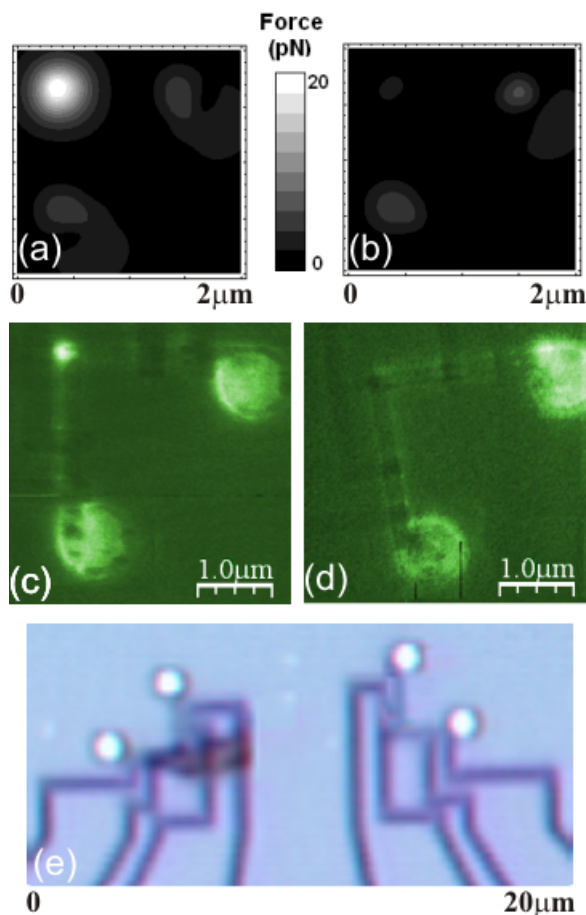


Figure 2. Simulation of the magnetic forces acting on a 130 nm Nanomag[®]-D bead at a distance of 200 nm from the device surface when the TDW has been created (a) and annihilated (b). Panels (c) and (d) report the corresponding MFM images, where bright contrast means an attractive tip-sample interaction. Panel (e): optical microscope image of two neighbouring devices on the same chip after a TDW has been injected only in the device on the left: the latter is the only one capable of attracting a cluster of beads (the dark region superposed on the leads).

As shown in figure 2(b), after the annihilation of the TDW the attractive force on the bead at the corner becomes negligible, whereas the force next to the disks remains essentially unchanged. On the disks the situation is similar to the previous case apart from a slight increase of the force in the upper right disk due to the slightly different spin configuration of the vortex in which the TDW is annihilated (see figure 1(b)).

These simulations are in good agreement with experimental magnetic force microscopy (MFM) images in figures 2(c) and (d). Indeed, there is an appreciable magnetic field generated in correspondence to the corner only when the TDW is there (figure 2(c)). After removal of the TDW there is some contrast only on the disks (figure 2(d)). The contrast is extended to the whole disk area, probably due to the interaction between the tip and the highly mobile vortex core. The analysis of the profile of the magnetic signal in figure 2(c) indicates that the maximum signal on the disk is about 1/4 of that on the corner.

The attraction properties of the device were verified in liquid through direct observation with an optical microscope. A

suspension of 130 nm Nanomag[®]-D streptavidin coated beads diluted in water (concentration of 1.1×10^{11} particles ml^{-1}) was used, and the device was capped with 150 nm of SiO_2 to avoid any interaction between the beads and the sensor material in the fluid. In every device we observed the process of attraction of beads by the TDW at the corner and their release upon annihilation of the TDW.

Figure 2(e) is an optical image of two devices on the same chip immersed in the suspension; the white circles correspond to the disks of figure 1(a). The TDW has been created only in the left device via the application and removal of a magnetic field along the bisector line of the left corner. Correspondingly a dark cluster of beads appears only in the left image, exactly where the TDW was created.

Theoretically, the disks are predicted to be effective in trapping some beads, but with a probability of attraction, which is at least ten times lower than the one at the corner (see figures 2(a) and (b)). However, in about 30% of the cases (about 50 experiments of capture under optical microscope), beads were observed to be trapped around the disk areas. This fact was also confirmed by an atomic force microscopy (AFM) study performed on the device, after evaporation of the water contained in the suspension of the same beads (concentration of 5×10^{10} particles ml^{-1}) previously dispensed on the surface with a pipette.

The high percentage of beads attracted by the disks is probably due to some imperfections in the device fabrication, producing an undesirable stray field on the contour of the disks and at the strip-disk connection. This would also explain the discrepancy between the values of the disk-to-corner force intensity ratio obtained in the simulation and measured in the MFM experiment.

We have verified, both theoretically and experimentally, that a further reduction of the strip width below 100 nm significantly reduces the field generated by the strip-disk connection. However the fabrication of such narrow structures (strip width of about 90 nm) on the Au contacts turned out to be more challenging and, so far, we have not succeeded in producing devices with an electric resistance, between the Au leads and the magnetic structure, low enough for the AMR measurement required in the detection process.

3. Detection experiments

Figure 3(a) illustrates the sensing concept for magnetic particle detection. As previously pointed out, the system switches from one micromagnetic configuration (figure 1(b)) to the other (figure 1(c)) at a defined value of the applied field. This field can be accurately evaluated by sensing the presence of the TDW at the corner through the AMR signal [16]. For this purpose, an AC current of 15 μA rms at 100 kHz was injected into contact 1, with contact 4 grounded, and the voltage drop between contacts 2 and 3 was measured through a lock-in amplifier.

The black curve in figure 3(e) represents the voltage drop measured across the corner of a clean device (before the beads have been dispensed). The TDW has been initially positioned at the corner and the magnetic field along one side of the

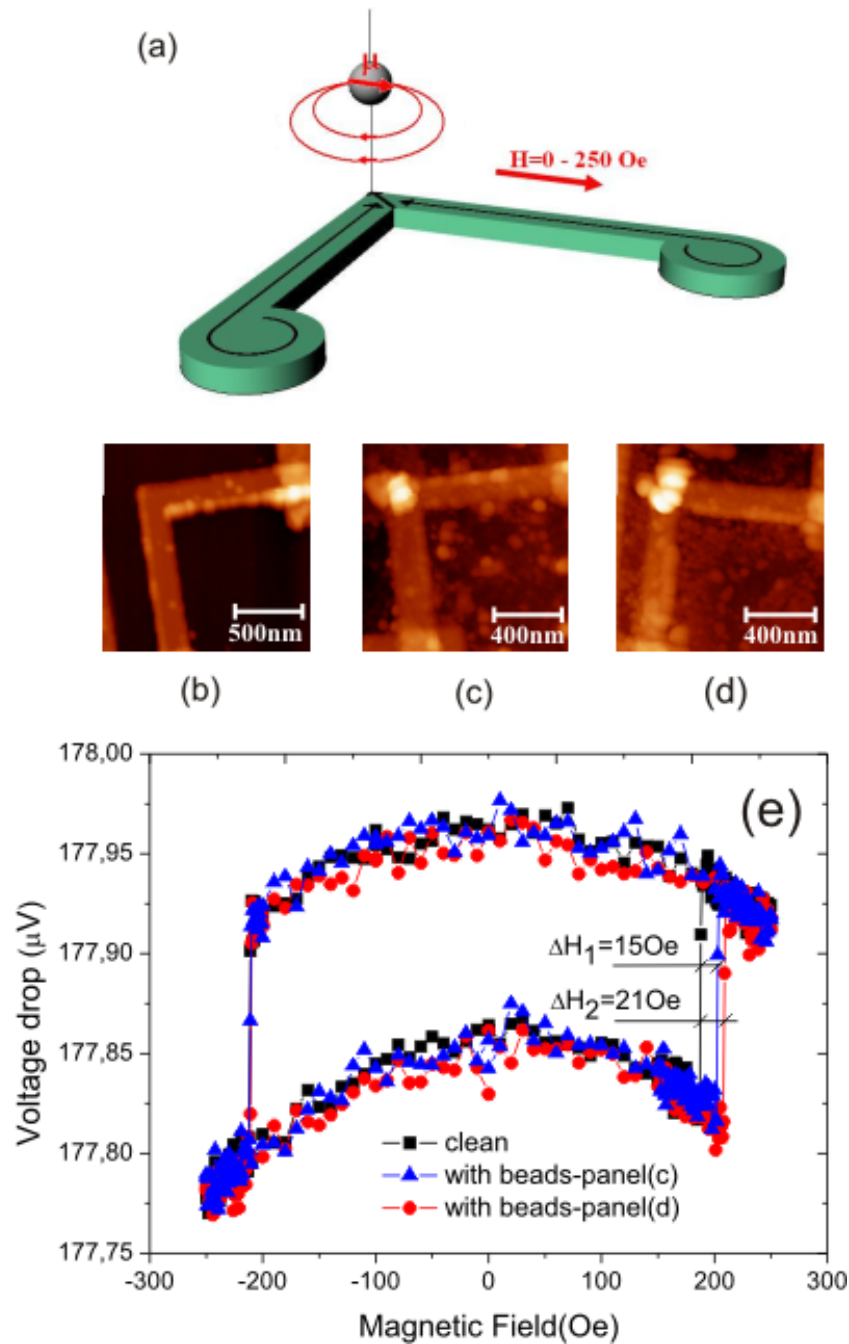


Figure 3. (a) 3D schematic of the device interacting with a magnetized particle. (b) AFM image of a clean device. (c) and (d) AFM images after the first and second dispensation of a drop of the same volume ($2 \mu l$) and concentration of beads (5×10^{10} particles ml^{-1}). From AFM analysis the cluster at the corner in (d) results as having a volume 1.5 times that of panel (c). (e) Voltage drop across the corner as a function of the magnetic field for the clean ring (squares), with the cluster of beads of panels (c) (triangles) and (d) (circles). The field is applied starting from 0 Oe and cycled back and forth up to ± 280 Oe.

corner has been increased from 0 to 280 Oe, then swept to -250 Oe, and finally brought back to the initial value. The resistivity of the permalloy is higher when the current and the magnetization direction are parallel, and lower when they are perpendicular ($\rho_{\parallel} > \rho_{\perp}$); this results in a different voltage drop depending on whether the TDW is placed at the corner or not. The transitions between the two configurations produce the two steps in the voltage signal. The first one, at positive field H_1 , corresponds to the annihilation of the TDW (resistance

increase) and the second one at a negative field H_2 (resistance decrease) indicates that the TDW is brought back to the initial position.

A preliminary characterization of more than 50 devices via AMR measurements led to an average value for H_1 and H_2 of 200 Oe and -220 Oe, respectively, with a variance of 15 Oe. However, for a given corner structure, the values of H_1 and H_2 are stable within ± 1 Oe, as checked in a sequence of loops (> 30) in which the TDW is brought back and forth to the corner.

It is also worthwhile observing that, by heating the devices up to 40 °C, the resistivity of the nanostructures changes at variance with the values of H_1 and H_2 . This result demonstrates the high stability of these devices with respect to temperature changes, at variance with other magnetoresistive sensors requiring accurate compensation or regulation of thermal drifts (Hall effects devices, GMR and TMR sensors).

When a bead is trapped at the corner by the TDW and an external field is applied to the device, the coupling between the induced dipole moment μ of the particle and the TDW below the particle causes an increase of the value of H_1 needed to annihilate the TDW, which can be revealed through the AMR signal, while H_2 is not affected.

Figure 3(b) shows an AFM image of a clean device (without capping layer) and figures 3(c) and (d) two cases when different amounts of beads are present on the same corner in two subsequent experiments. The results of the magnetoresistance measurements in the three cases are displayed in figure 3(e), where the black, blue and red curve correspond to figures 3(b), (c), (d), respectively.

In these experiments we used streptavidin coated MICROMOD Nanomag[®]-D beads diluted in NH_4OH with a concentration of 5×10^{10} particles ml^{-1} in order to avoid clustering. For the case in figure 3(c), 2 μl of suspension has been initially dispensed on the device with a pipette and the measurements (AFM and magnetoresistance) were performed after evaporation of the solvent. The procedure was repeated a second time on the same device, thus leading to an additional accumulation of particles visible in figure 3(d).

The beads grouped on the corner by the TDW can only influence the position of the first transition (for positive magnetic field) corresponding to the removal of the TDW from the corner where the beads are located, while the second step (for negative fields) is unaffected by the presence of beads on the corner because it depends on the creation and propagation of a TDW from the region of the terminating disks. The second step can then be used as a reference in all the other measurements, in order to avoid erroneous evaluation of the TDW depinning field (DF) due to angular misalignment or imperfect control of the applied field.

The presence of the cluster in figure 3(c) causes an increase of the DF by 15 Oe, while the cluster in figure 3(d) produces an increase of 21 Oe. Note that these variations are largely above the uncertainty in the DF (± 1 Oe). Counting the exact number of beads present on the corner in the two experiments turned out to be particularly difficult, mainly because these beads are stable only in water; when dried they become a rough mixture of magnetic nanoparticles and portions of the polymeric matrix where the magnetic nanoparticles are dispersed. An estimate of the volume of material present on the device surface was obtained with AFM; for the cases reported, the volume of the beads in figure 3(d) is $7 \times 10^6 \text{ nm}^3$, nearly 1.5 times the volume of the cluster in figure 3(c).

In any case it is very difficult to correlate the variation of the DF with a reliable value of the magnetic moment of the particles on top of the corner, due to the unknown mixture of magnetic material and polymer in the clusters seen

by AFM after drying. In addition, as was recently pointed out from analytical calculations [17], the average magnetic field produced by a single bead close to a sensor is strongly dependent on bead position with respect to the sensing area. Therefore the exact position of the magnetic particles inside the cluster, with respect to the corner, is another crucial variable determining the device response.

In order to better characterize the response of the device, for different thicknesses of an SiO_2 capping layer, synthetic antiferromagnetic (SAF) nanoparticles [18] were also employed.

These particles are realized using nanoimprint lithography on a ferromagnetic multilayer structure, followed by releasing and stabilization in solution. They are produced in the form of disks, with a 70 nm diameter and 20 nm thickness; made of two CoFe layers separated by a Ru spacer and antiferromagnetically coupled via bilinear coupling. In the multilayer structure the interlayer magnetic interactions are exploited in order to produce particles with zero remanence and a quasi-superparamagnetic behaviour. The main reason for their use in this work is the absence of a polymeric matrix, giving us the possibility to estimate their number by measuring the total volumes on the corner.

In addition, SAF particles present a high magnetic moment at saturation μ_s , which for the single particle is estimated to be of the order of 10^{-13} emu, from measurements with an alternating gradient magnetometer (AGM) on 5 μl of a suspension of particles. This value is comparable to the μ_s of the 130 nm beads previously used, even if the volume of the single particle is more than 20 times smaller. The particles were diluted in NH_4OH , to a final concentration of 2.7×10^9 particles ml^{-1} , in order to get the minimum amount of nanoparticles on the corner at every dispensation of 1 μl of suspension (particles were provided in a concentration of 2.7×10^{11} particles ml^{-1}).

The AFM analysis, performed after the drop evaporation, allowed the determination of the volume of the clusters on the corner. In the following we will compare situations where the number of SAF nanoparticles attracted by the corner corresponds to 6–8 particles, independent of the presence and thickness of the SiO_2 capping layer.

The graph of figure 4 shows the DF variation as a function of the thicknesses of the SiO_2 , measured on different devices, for a typical cluster size on the corner mentioned above (see the inset of figure 4). The graph shows that the DF variation decreases as the capping layer thickness increases. As the maximum error in the measurement of DF is ± 1 Oe, the graph of figure 4 indicates that the detection of a single SAF nanoparticle is possible for capping layer thicknesses below 25 nm.

At a 50 nm thickness of the capping layer, the switching field variation *per bead* is of the same order of magnitude as the error in the evaluation of the DF, thus the detection of a single SAF nanoparticle is not achievable.

With a SiO_2 thickness of 120 nm the average measured change of the DF was 3 Oe and this corresponds to the limit of detection for the typical cluster size considered.

Additional measurements, using Nanomag[®]-D beads of 300 and 500 nm diameter, show that a single bead can be

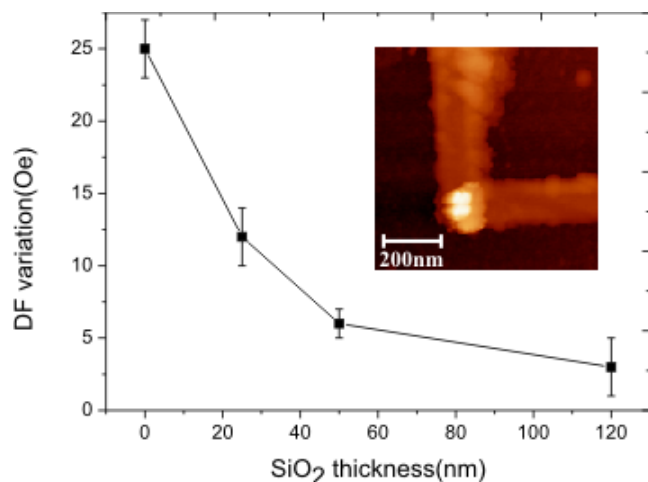


Figure 4. Measured depinning field variation as a function of the thickness of the SiO₂ capping layer. The error bar indicates the variation in the DF for different measurements in the same conditions. The inset (800 nm × 800 nm) shows the typical cluster of SAF nanoparticles found at the corner of the device after dispensation of 1 μ l of suspension.

detected even with a 120 nm capping layer, the DF variation per bead being equal to 8 and 12 Oe, respectively.

In order to investigate the operation of the device in a liquid environment we also performed preliminary investigations using 500 nm beads. The results obtained so far indicate the possibility of detecting beads in suspension, i.e. without waiting until the drop has evaporated as in the previous experiments. We measured, with good reproducibility, a DF variation of the order of 10–15 Oe one minute after the deposition of the drop, and before complete evaporation. Optical inspection of the samples, after the bead detection, confirmed that the observed DF variation was indeed related to the capture of beads by the corner of those devices where a TDW is present. A more systematic study on the detection of beads in suspension is on the way, requiring the optimization of the set-up and the development of a fluidic system on top of the sensor in order to guarantee more reproducible conditions of bead dispensation and avoid the evaporation of the solution.

4. Conclusion

In summary, a device, capable of acting as a switchable magnetic pole for attracting and detecting magnetic beads in suspension on its sensing area, has been presented.

The attraction properties of the corner and disks that compose the device were studied both theoretically and experimentally, showing that beads are mainly attracted where the transverse domain wall lies. The detection of a single SAF magnetic nanoparticle (disk with diameter of 70 nm and thickness of 20 nm) for protective capping layer thickness of up to 25 nm was demonstrated.

For a thicker capping layer (120 nm) the detection of a single bead of larger size (300 and 500 nm) was also

demonstrated. Our device has the potential for application as a bead counter in microfluidics systems or potentially as a sensor for single molecule detection if the active area is properly functionalized with the specific probe molecule.

Acknowledgments

This work was partially funded by Fondazione Cariplo via the project SpinBioMed (Project No. 2008.2330). PV acknowledges funding from the Department of Industry, Trade, and Tourism of the Basque Government, the Provincial Council Gipuzkoa under the ETORTEK Programme (Project No. IE06-172), the Spanish MICINN (Project CSD2006-53), and the EU VII Framework Programme (grant agreement No. PIEF-GA-2008-220166). VM acknowledges support by the US NSF, Grant ECCS-0823813 and by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357 (CNM ANL grants Nr.468 and Nr.470). MZ and SXW wish to thank support from a Stanford Graduate Fellowship and National Cancer Institute Grant 1U54CA119367 (CCNE-TR).

References

- [1] Pankhurst Q A, Connolly J, Jones S K and Dobson J 2003 *J. Phys. D: Appl. Phys.* **36** R167
- [2] Chiou C H, Huang Y Y, Chiang M H, Lee H H and Lee G B 2006 *Nanotechnology* **17** 1217
- [3] Dobson J 2008 *Nat. Nanotechnol.* **3** 139
- [4] Graham D L, Ferreira A H and Freitas P P 2004 *Trends Biotechnol.* **22** 455
- [5] Li G, Joshi V, White R L, Wang S X, Kemp T X, Webb C, Davis R W and Sun S 2003 *J. Appl. Phys.* **93** 7557
- [6] Shen W, Schrag B D, Carter M J, Xie J, Xu C, Sun S and Xiao G 2008 *J. Appl. Phys.* **103** 07A306
- [7] Besse P A, Boero G, Demierre M, Pott V and Popovic R 2002 *Appl. Phys. Lett.* **80** 4199
- [8] Ejsing L, Hansen M F, Menon A K, Ferreira H A, Graham D L and Freitas P P 2004 *Appl. Phys. Lett.* **84** 4729
- [9] Lee C S, Lee H and Westervelt R M 2001 *Appl. Phys. Lett.* **79** 3308
- [10] Janssen X J A, van IJzendoorn L J and Prins M W J 2007 *Biosens. Bioelectron.* **23** 833
- [11] Jiang Z, Llandro J, Mitrelias T and Bland J A C 2006 *J. Appl. Phys.* **88** 08S105
- [12] Mihajlović J, Xiong P, von Molnár S, Field M and Sullivan G J 2007 *J. Appl. Phys.* **102** 034506
- [13] Reiss G 2008 private communication
- [14] Vavassori P, Metlushko V, Ilic B, Gobbi M, Donolato M, Cantoni M and Bertacco R 2008 *Appl. Phys. Lett.* **93** 203502
- [15] Donahue M J and Porter D G 1999 *OOMMF User's Guide, Version 1.0. Interagency Report NISTIR 6376* (Gaithersburg: National Institute of Standards and Technology)
- [16] Vavassori P, Metlushko V and Ilic B 2007 *Appl. Phys. Lett.* **91** 093114
- [17] Damsgaard C D and Hansen M F 2008 *J. Appl. Phys.* **103** 064512
- [18] Hu W *et al* 2008 *Adv. Mater.* **20** 1479