Domain wall displacement in Py square ring for single nanometric magnetic bead detection

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An approach based on domain wall displacement in confined ferromagnetic nanostructures for attracting and sensing a single nanometric magnetic particle is presented. We modeled and experimentally demonstrated the viability of the approach using an anisotropic magnetoresistance device made by a micron-sized square ring of Permalloy. This detection concept can be suitable to biomolecular recognition and, in particular, to single molecule detection. © 2008 American Institute of Physics. [DOI: 10.1063/1.3030984]

Intense research is presently directed toward the development of high sensitivity magnetoresistive sensors for the detection of magnetic beads because of their potential high impact on biochemical applications and diagnosis in medicine. Since the pioneering work by Baselt et al.,1 magnetic beads have been used for labeling and detecting target molecules at the surface of magnetic sensors where probe molecules are immobilized. High biological sensitivity can be achieved in immunoassays, theoretically down to the single molecule detection if nanobeads are employed. This is particularly appealing for biomedical research, offering the opportunity of immobilizing and detecting a biomolecule at a desired location for fundamental studies of its functions and dynamics.2 A number of very sensitive magnetic field detection devices have been developed during recent years, which are able to detect a single micromagnetic particle, such as giant magnetoresistance (GMR) sensors,3 spin-valves,3 miniaturized silicon Hall sensors,4 planar Hall effect sensors based on Permalloy thin films,5 and tunneling magnetoresistance sensors.6 The use of the anisotropic magnetoresistance (AMR) effect in ring-shaped sensors as sensitive bead detectors was suggested by Miller et al.,7 and more recently the approach has been extended by Llandro et al.,8 to ring-shaped multilayered (pseudo-spin-valve) sensors based on GMR. In some cases the possibility of detecting single nanometric beads has been foreseen.9 However, to date, none of these approaches has experimentally demonstrated recognition of single nanometric magnetic beads with a diameter below 100 nm, particularly appealing for reducing the perturbation on the affinity between target and probe molecules.

Here we demonstrate the capability of detection of individual nanometric magnetic beads with a diameter of 80 nm previously attracted to the active sensing area of an AMR sensor based on a ferromagnetic micron-sized square ring. A salient feature of the sensor concept presented here10 is that the entire device does not need to be as small as the nanobeads to be sensed, making its fabrication less challenging. In fact the nanometric size of the sensing area arises from the physical properties of a domain wall (DW) localized at a geometric corner. The concept illustrated in the present paper relies on a previous experimental work made on square rings of Permalloy (Py) for application in magnetic storage of information.11 In this design head-to-head and tail-to-tail DWs having a transverse structure (Néel type)11 can be positioned at a given corner, and their position can be read electrically thanks to the AMR effect: when a DW is present between two sensing leads a reduction in the resistance is observed since some of the magnetization of the DW points perpendicularly to the current flow. Otherwise, when there is no DW present between the two sensing leads, the magnetization follows the direction of the perimeter of the ring and the resistance is higher. In this work we adapted this device to demonstrate a detection concept suitable for the detection of magnetic nanobeads. Panel (a) of Fig. 1 shows the scanning electron microscopy image of the structure used in the present experiment. The 30 nm thick Py square ring structures have been lithographically patterned on top of 20 nm thick and 100 nm wide Au contacts, previously fabricated on a SiO2/Si substrate. The outside size of the rings is 1.0 × 1.0 μm2, the width of each segment is about 180 nm, and the slit is about 80 nm wide. For the magnetoresistance measurements presented here, the voltage drop was measured using a lock-in amplifier between contacts labeled 3 and 4 in the panel (a) of Fig. 1, with a current of 15 μA injected at contact 1 while contact 2 was grounded. The three-dimensional (3D) schematic shown in panel (b) of Fig. 1 illustrates the sensing concept proposed here: when a bead is placed over a DW previously positioned at one corner of the ring structure and a magnetic field H is applied to displace the DW along one ring edge, a magnetic dipole moment μ is generated in the superparamagnetic bead, as shown in Fig. 1. The stray field generated by μ opposes the applied field below the bead, causing an increase in the value of the field H required to displace the DW. In panel (c) of Fig. 1 we present the result of a simulation of the effect when a magnetic nanobead (a commercial MICROMOD nanomag®-D with diameter 130 nm and magnetic moment μ = 150 × 10−15 emu at 1000 Oe) is placed over the DW at a vertical distance of 15 nm from the surface of the Py structure. 3D micromagnetic
simulations of the whole system were performed by using the object oriented micromagnetic framework (OOMMF). In detail, the graphs in Fig. 1c show the variation in the magnetization component parallel to the applied field, normalized to its saturation value in the upper segment of the ring structure. Panel a of Fig. 1 shows the simulation of the device with the magnetic force microscopy (MFM) image in panel c of Fig. 2, showing that there is a magnetic field emanating from the structure only in correspondence of the DWs (left-top and bottom-right corners in the image). Elsewhere the field is negligible. The strong field gradient that characterizes this stray field causes a self-focusing action that can trap and drag toward the corner sensing region a nanoparticle flowing in the vicinity of the structure. The force acting on a nanoparticle at a certain distance from the ring surface can be calculated by computing with OOMMF the magnetic field H created in the surrounding space by the nanostructure in the magnetic configuration with a DW at the two opposite corners of the ring. Then the following vector expression is used for the force: \( \mathbf{F} = -\mu_0(\mathbf{\mu} \cdot \nabla)\mathbf{H} \), where \( \mathbf{\mu} = \mu(\mathbf{H})\mathbf{h} \) with \( \mu(\mathbf{H}) \) the known magnetization curve of the bead and \( \mathbf{h} \) is a unit vector parallel to the field \( \mathbf{H} \).

Figure 3 shows the results of the magnetoresistance measurements carried out to verify the sensing concept presented here onto rings without SiO\(_2\) capping. Panel (a) shows the AFM image taken from a clean ring, while panel (b) shows the AFM image of the same ring after the dispensation of beads of 130 nm diameter (MICROMOD nanomag\(^{\circledD}\)-D) in ethanol and rinsing with de-ionized water for 1 min. In panel (b) the white line indicates the position of the DW in the corner of the ring and subsequently dispensed the beads on the chip surface. The latter was capped with a 30 nm thick SiO\(_2\) layer to avoid any local specific chemical interaction between beads and the different materials of the sensors. We typically dispensed a 1 \( \mu l \) drop in solution of beads in ethanol with a final concentration of about \( 10^6 \) particles/\( \mu l \) and then immediately dried the surface with nitrogen. We employed two types of commercial nanobeads, MACSTM and nanomag\(^{\circledD}\)-D, having diameters of 50 and 130 nm. The AFM image presented in panel (a) refers to the 50 nm beads and shows that a cluster of several beads is formed over one of the corners where a DW is located. The AFM image in panel (b) shows a single 130 nm bead placed over the opposite corner, where the other DW is located. We repeated the experiment several times changing either ring structure or the DW position and we always observed that clusters (most common for the 50 nm beads) or single particles are found over the corners where the DWs are located, but never over the other two corners of the ring. This interesting behavior can be understood by looking at the magnetic force microscopy (MFM) image in panel (c) of Fig. 2, showing that there is a magnetic field emanating from the structure only in correspondence of the DWs (left-top and bottom-right corners in the image). Elsewhere the field is negligible. The strong field gradient that characterizes this stray field causes a self-focusing action that can trap and drag toward the corner sensing region a nanoparticle flowing in the vicinity of the structure. The force acting on a nanoparticle at a certain distance from the ring surface can be calculated by computing with OOMMF the magnetic field \( \mathbf{H} \) created in the surrounding space by the nanostructure in the magnetic configuration with a DW at the two opposite corners of the ring. Then the following vector expression is used for the force: \( \mathbf{F} = -\mu_0(\mathbf{\mu} \cdot \nabla)\mathbf{H} \), where \( \mathbf{\mu} = \mu(\mathbf{H})\mathbf{h} \) with \( \mu(\mathbf{H}) \) the known magnetization curve of the bead and \( \mathbf{h} \) is a unit vector parallel to the field \( \mathbf{H} \). Panel (d) of Fig. 2 shows the contour plot of the modulus of the attractive force acting on a nanobead of 130 nm in diameter computed in a plane at 200 nm distance from the sensor surface. The plot shows that a force in the range of 0.1–10 pN is acting on the bead over an area of about 400 nm in diameter, ensuring an effective trapping and focusing action on the nanobead.

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measurements. Panel face took place, keeping the beads at a fixed position during chemical interaction between the beads and the sensor superimposed over and next to the corner. Fragmentation revealed to smaller beads, having an average diameter of about 80 nm, top-left corner of the ring structure, while the circles evident to the displacement field of the order of 12 Oe in agreement with the experimental value. This is obtained for the minimum beads-sensor distance compatible with the mesh (15 nm), while this value is unaltered when increasing the distance up to 30 nm. Simulations also indicate that only the two beads over the DW have an influence, with the bead exactly on top of the DW (indicated by the arrow in Fig. 3) that is essentially responsible of the shift of 12 Oe, while the other one gives a contribution of only 2 Oe. In this sense our experiment demonstrates the detection of two beads, and also the possibility of single bead detection as the error in the evaluation of the displacement field is ±2 Oe. Moreover simulations show that the reduction in the width of the ring to the nanobead diameter doubles the value of ΔH. This in conjunction with the use of nanobeads of a higher magnetic moment (values up to 5 times that of the beads used here are reported in the literature) can increase the achievable value of ΔH by about ten times.

To summarize we demonstrated the viability of an approach for sensing single nanometric beads based on their impact on the displacement field of a DW at a corner of a Py nanostructure. The self-focusing effect on the active sensor area due to the strong magnetic field emanating from the DW has also been demonstrated. This proof of concept paves the way to the optimization of the sensor geometry for improving sensitivity and fulfilling the requirements imposed by biological recognition in view of application to single biomolecule detection and localization.

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