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Using permalloy based planar hall effect sensors to capture and detect superparamagnetic beads for lab on a chip applications

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ABSTRACT

Experimental studies have been carried out on planar Hall effect (PHE) sensors used to detect magnetic nanoparticles employed as labels for biodetection applications. Disk shaped sensors, 1 mm diameter, were structured on Permalloy film, 20 nm thick. To control the sensor magnetisation state and thus the field sensitivity and linearity, a DC biasing field has been applied parallel to the driving current. Maghemite nanoparticles (10 nm) functionalised with Polyethylene glycol (PEG) 6000 were immobilised over the sensor surface using particular magnetisation state and applied magnetic fields. In order to obtain a higher response from the magnetic nanoparticles, it was used a detection setup which allows the application of magnetic fields larger than 100 Oe but avoiding saturation of the PHE signal. Based on this setup, two field scanning methods are presented in this paper. During our experiments, low magnetic moments, of about 1.87×10^{-5} emu, have been easily detected. This value corresponds to a mass of 9.35 μg of maghemite nanoparticles functionalised with PEG 6000. The results suggest that this type of structure is feasible for building low cost micrometer sized PHE sensors to be used for high-resolution bio sensing applications.

1. Introduction

Superparamagnetic (SPM) micro- and nanobeads are versatile tools in lab-on-a-chip (LOC) applications. Because of their magnetic properties and small dimensions, they offer possibilities to label, actuate, and detect with high sensitivity chemical and biological species. The magnetite or maghemite beads, which are among the most used for LOC applications, show superparamagnetic behavior above the blocking temperature, T_B, i.e., the measured magnetic moment in the absence of an external magnetic field is zero. An external magnetic field can magnetize the nanobeads, like a paramagnet but their magnetic susceptibility is much larger than the one of paramagnets and the saturation effect appears for large applied fields. This SPM behavior is useful in LOC applications because can be avoided unwanted capture of the beads over the sensors surface and can be minimised false detection signals when magnetic sensors are used. From ZFC-FC magnetisation measurements made on maghemite nanoparticles, 10 nm in diameter, we have obtained $T_B = 252$ K.

Magnetic sensors, based on giant magnetoresistance (GMR), tunneling magnetoresistance (TMR) [1,2] or planar Hall effect (PHE) [3–5], can be integrated in biochip platforms for measuring the fringe fields created by SPM beads used as labels for different biomolecular targets. In fact, the presence of beads at the sensor surface forms the basis for most SPM bead sensing platforms. It should be noted that for this detection setup it has been observed, both by experiments and micromagnetic simulations, a signal dependence on the spatial location of magnetic nanoparticles over the sensor surface [6–8]. This issue becomes very important when a small number of biomolecules must be detected using the surface immobilisation method because the signal output of the sensor may experience large variations depending on the position of the beads.

Finally, we should mention that the strong localized stray field from domain walls (DWs) in sub micrometer ferromagnetic tracks can trap individual SPM beads with forces up to hundreds of pN and manipulate them [9]. So, a strong magnetostatic interaction between the magnetic beads and the sensor surface can appear and be responsible not only for the actuation and capturing of magnetic beads over the sensor surface but, also, for complex changing of the magnetic moments orientation in the sensing layer due to the stray field produced by the beads.

Based on these findings we investigate the behavior of Permalloy based PHE sensor disks under different applied fields in order to find the conditions for which the magnetostatic interaction between the SPM beads and the sensor surface become large

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enough to capture the beads inside of the sensor surface and, then, to detect their presence. We show that very simple and low cost structures can offer high detection sensitivity, which can be lower than 10^{-5} emu, and the possibility to capture SPM nanobeads over the sensor surface.

In a previous study, [7], the chip with PHE sensors was mounted on a soft magnetic grid. A constant biasing field was applied in the film plane, parallel with the driving current through the sensor, and then removed before making measurements. The signal dependence on beads position over the sensor surface has been studied. Now, the chip with the same design of the PHE sensors is placed on a non magnetic grid. By this, the magnetization state of the PHE sensors can be precisely controlled using both in plane and perpendicular applied fields allowing flexibility in setting up various detection methods as we will show in this paper. In addition, we expect an increasing of sensor's output dynamic compared to the case when the chip was mounted on a magnetic grid. This behavior was evidenced by our micromagnetic simulations presented in the previous study i.e., a larger difference between the PHE signals with and without nanobeads over the sensor surface.

2. Experiments

A Permalloy film, 20 nm thick, has been deposited on to oxidised Si substrate. No magnetic anisotropy axis has been defined during the deposition. Disk-shaped PHE sensors, 1 mm diameter, were structured on the Permalloy film by using photolithography technique. Also, have been patterned Au pads, 250 μm length, that are in contact with the Permalloy disks to define the PHE structures, Fig. 1(a), [7]. The sensors were passivated by sputtering a 200 nm thick TiO_2 layer to protect them against the fluid used during the experiments. In Fig. 1(a) are illustrated the electrical connections setup, the directions of the applied, H_{appl} and biasing, H_{bias}, fields respectively. This is typical setup used for field sensing. Two detection sites have been defined on the chip by using pairs of measurement and reference sensors. The chip was mounted on a custom designed non magnetic grid, made from glass-reinforced epoxy printed circuit board. The chip with PHE sensors was placed in a home-made system composed of Helmholtz coils which are able to generate well defined and uniform magnetic fields over three orthogonal directions; two of these fields are generated in the film plane. The measurement system, used to study the field dependences of the AMR and PHE effects, consists in Keithley 6221 programmable current source, Keithley 2182 A nanovoltmeter, and three programmable high current sources. The DC driving current through the sensors, $I_{sens}=5$ mA, was chosen to maximize the output signal but without affecting the thermal stability of the structure. The resistance between the current or the voltage contacts is about 120 Ω. A DC detection setup was used to read out the sensor because the frequency of the sweeping magnetic field was 0.01 Hz. The integration time, i.e. the period of time the input signal is measured, was 20 ms. In addition, a digital filter has been used. For each displayed reading, five measurements were averaged. This offers the best compromise between noise performance and speed. For this setting, the noise level was about 35 nV.

The magnetization curves for functionalised maghemite nanoparticles with Polyethylene glycol (PEG) 6000 were measured at room temperature with 7 T Mini Cryogen Free Measurement System from Cryogenic.

3. Results and discussion

The PHE is due to the anisotropic magnetoresistance (AMR) effect found in magnetic materials. A study presented in [10] clearly illustrates the field dependence of the AMR effect using a typical Hall effect measurement setup which is, from electrical point of view, similar to a Wheatstone bridge. In such experiments the field is applied in the film plane. It was found a quadratic dependence of the PHE signal on the magnetization, M, in Ni, Co, Fe, and Ni_xFe_{1-x} films. The output signal also shows an angular dependence such that a general equation, of the type [7] $U_{PHE}\sim I_{sens}M^2\sin 2\theta$, can be used to describe the PHE signal; θ is the angle between the magnetization vector and the driving current through the sensor, I_{sens}. In turn, M depends on the applied field and the signal can be used as a probe of the structure magnetisation.

In a previous study [7] we presented the AMR curves measured for the sensors, S1 and S2, placed on the chip and connected like in Fig. 1(a). The applied field, H_{appl}, makes the angles 45° (for S1) and 135° (for S2) with the driving current and $H_{bias}=0$; in this case $\sin 2\theta=\pm 1$. The AMR effect saturates for fields higher than 50 Oe which means that the sensors cannot be used for detection of in plane applied fields higher than 25–50 Oe. The magnetization curve [7] of the four sensors, placed on the chip, shows a very small hysteresis effect. We used a simple method to find with precision the coercive field by applying the biasing field parallel to the driving current through the sensor S1, like in Fig. 1(a), and to sweep this field between ±150 Oe; $H_{appl}=0$. In this way, the film magnetisation will be parallel or antiparallel with the sensor

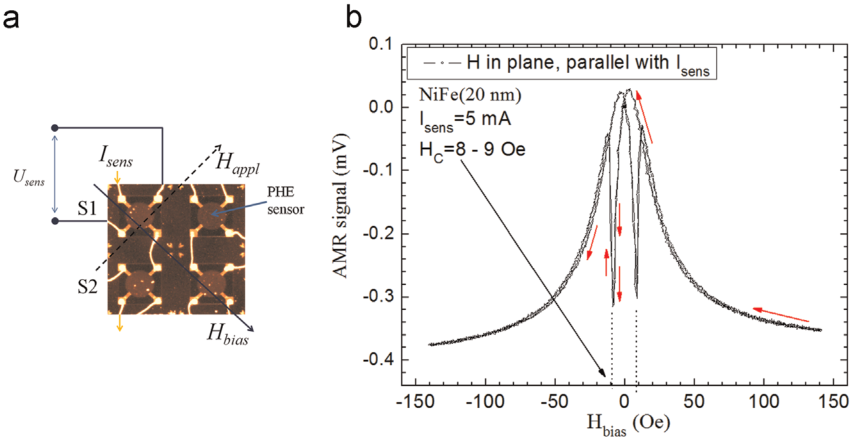

Fig. 1. (a) Enlarged top view of the chip with 4 PHE sensors (adapted with permission from M. Volmer and M. Avram, Signal dependence on magnetic nanoparticles position over a planar Hall effect biosensor, Microelectron. Eng. 108 (2013) 116–120, Elsevier). The biasing field, H_{bias}, and the applied field, H_{appl}, directions are illustrated. Also, the electrical connections are indicated; (b) The AMR signal measured when the biasing field is swept between ±150 Oe. The arrows are guides for the eyes.