Bead Capture on Magnetic Sensors in a Microfluidic System

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Abstract—The accumulation of magnetic beads by gravitational sedimentation and magnetic capture on a planar Hall-effect sensor integrated in a microfluidic channel is studied systematically as a function of the bead concentration, the fluid flow rate, and the sensor bias current. It is demonstrated that the sedimentation flux is proportional to the bead concentration and has a power law relation to the fluid flow rate. The mechanisms for the bead accumulation are investigated and it is found that gravitational sedimentation dominates the bead accumulation, whereas the stability of the sedimented beads against the fluid flow is defined by the localized magnetic fields from the sensor.

Index Terms—AMR, bead, biosensor, lab-on-a-chip, microfluidic.

I. INTRODUCTION

MAGNETIC biosensors relying on magnetic detection of magnetic beads bound to the sensor surface by a bioassay show great potential for being compact biosensors with a direct electrical readout (for reviews, see [1]–[5]). Several magnetic sensor principles are being explored, e.g., giant magnetoresistance and spin valve sensors [3]–[7], magnetic tunnel junctions [3], [4], micro-Hall sensors [8], and planar Hall effect (PHE) sensors [9]–[11].

Common for all sensor principles is that the biological sample and the magnetic bead suspension is introduced to the sensor followed by washing steps. To achieve reproducible conditions under these various incubation and washing steps, it is desirable to integrate the sensor in a fluidic system. To design this system, it is important to know how the experimental conditions influence the results. Previously, the sedimentation of beads from a stagnant bead suspension on a Wheatstone configuration of spin-valve sensors in a trench has been studied by Thilwind et al. [7].

In this study, we focus on the interaction between magnetic beads in a flowing suspension and the sensor surface of PHE sensors. Using the electromagnetic field from the bias current passed through the sensor, we systematically study the real-time response of PHE sensors integrated in a microfluidic channel exposed to a bead suspension as function of the time $t$, the bead concentration $c$, the fluid flow rate $Q$ and the sensor bias current $I_{\text{bias}}$. The experimental studies are supplemented by theoretical calculations and a discussion of the bead accumulation mechanisms.

II. EXPERIMENTAL

A. System Design and Fabrication

The exchange-biased PHE sensors with the structure Si/SiO$_2$(800 nm)/Ta(5 nm)/Ni$_8$Fe$_{12}$O$_{30}$ nm)/Mn$_{27}$Ir$_{24}$(20 nm)/Ta(5 nm) were deposited by ion beam deposition as described in [12] and subsequently defined by ion milling. During deposition, a magnetic flux density of 4 mT was applied to define an easy magnetization direction of the permalloy layer in the positive $x$-direction (see Fig. 1). Gold contacts were deposited by e-beam evaporation and defined by liftoff. Subsequently, a 470 nm protective layer of SiO$_2$ was RF sputter deposited through a shadow mask. A sensor chip had three sensor crosses that each had an active area of $w \times w = 40 \mu m \times 40 \mu m$. The chip was mounted on and wire bonded to a ceramic chip carrier. During experiments, a current $I_x$ was applied through the sensors in the $x$-direction and the voltage drop $V_y$ along the $y$-direction was measured as indicated for the middle sensor.
in Fig. 1. For low fields $H_y$ along the $y$-direction, the sensor output is

$$V_y = I_x S_0 H_y$$  \hspace{1cm} (1)

where $S_0$ is the sensor sensitivity [10]. The sensitivity for the sensors used in the experiments was found to be $6.2 \times 10^{-5}$ V nT$^{-1}$.

A fluid system was assembled around the sensor and substrate as sketched in Fig. 1. The fluid system was defined by laser cutting in PMMA and consisted of a 2-mm-thick fluid plate defining the fluid chamber glued to the ceramic chip carrier and a top with fluid inlets and outlet. The channels on the top part were sealed by lamination foil. The wire bonds were protected with silicone glue (Elastosil E41, Wacker Chemie, München, Germany). This glue also sealed the interface between the ceramic chip carrier and the fluid chamber plate. The two-part fluid system was sealed by an O-ring. The channel cross section at the chip was $w_c \times h_c = 6 \text{ mm} \times 1.1 \text{ mm}$. The system had two inlets to allow for an easy switching between bead suspension and water.

To ensure a constant temperature during experiments, the system was mounted on a Cu block with integrated thermometer and temperature control by use of a Peltier element. Fig. 2 shows the system mounted in the experimental setup with temperature control.

B. Experimental Procedure

The two fluid inlets were connected to syringes with MyOne bead suspension and deionized (DI) water, respectively. The fluid flow rates were controlled by Harvard Apparatus PHD 2000 syringe pumps. An ac sensor bias current with a frequency of 2200 Hz was supplied by a Keithley 6221 current source. After preamplification in a Stanford Research Systems (SRS) SR552 bipolar preamplifier, the signal was extracted by use of a SRS SR830 lock-in amplifier. During all experiments, the sample holder temperature was kept constant at 28 °C. All measurements were carried out in zero applied magnetic field and without any magnetic shielding.

All experiments and calculations were carried out with MyOne beads, which have a diameter of $D = 1.05 \mu$m, a density of $\rho_b = 1700$ kg/m$^3$ and a measured magnetic susceptibility of $\chi = 1.377$ [13].

Prior to experiments, the fluid system was flushed with DI water to remove air bubbles. Then, a syringe with freshly mixed bead suspension of mass-to-volume concentration $c$ was mounted in the syringe pump and the bead suspension was injected at a flow rate $Q$. The responses of all three sensors were monitored versus the time $t$ after the bead suspension entered the fluid system. Subsequently, the fluid system was cleaned by letting an air bubble into the channel followed by rinse with DI water. It was verified that the sensor signals returned to their baseline level after washing. A comparison of the signals from the three sensors as well as repeated experiments verified that the experimental observations were reproducible.

III. THEORY

A. Sensor Response to Self-Field

We consider the sensor response for an ac applied current

$$I_x(t) = I_x0 \sin(\omega t)$$  \hspace{1cm} (2)

in zero external magnetic field. In the absence of magnetic beads, the sensor is still affected by an average field in the $y$-direction because part of the applied current is shunted in the antiferromagnetic Mn$_7$Co$_{13}$Fe$_{23}$ layer. We write this field as $\gamma_0 I_x$, where $\gamma_0$ is a constant related to the sensor stack and geometry. When the antiferromagnet is deposited on top of the permalloy layer, the field from a current passing through the antiferromagnetic layer in the positive $x$-direction gives rise to a field acting on the ferromagnetic layer in the positive $y$-direction and hence $\gamma_0$ is positive. When beads are present on the sensor, they will also give rise to a positive field in the $y$-direction, which we write as $\gamma_1 I_x$ [14]. Hence, the sensor signal due to its self-magnetization and beads can be written as

$$V_y = I_x^2 \gamma_0 \sin^2(\omega t) S_0 (\gamma_0 + \gamma_1).$$  \hspace{1cm} (3)

The signal is detected by a lock-in amplifier. The lock-in amplifier multiplies $V_y$ with a reference signal $V_{ref}(t) = \sqrt{2} \sin(n\omega t + \phi)$, where $n$ is the harmonic chosen for the detection and $\phi$ is the phase of the reference signal ($\phi = 0$ for detection of the in-phase response and $\phi = \pi/2$ for detection of the out-of-phase response). Subsequently, the signal is passed through a low-pass filter. The measurements in the present study are all carried out using the second harmonic out-of-phase response ($n = 2$, $\phi = \pi/2$), which is calculated to

$$V_{y2}'' = -\frac{1}{2\sqrt{2}} S_0 I_x^2 \gamma_0 (\gamma_0 + \gamma_1).$$  \hspace{1cm} (4)

Thus, assuming that $\gamma_1$ is proportional to the bead coverage, we can use measurements of $V_{y2}''$ to monitor the amount of beads in the vicinity of the sensor.

B. Bead Sedimentation

We consider the motion of a bead in a fluid flow under the influence of buoyancy, the Stokes fluid drag force and magnetic forces. The equation of motion for a single bead with the coordinate system defined in Fig. 1 is

$$\mathbf{F}_{\text{bead}} = \mathbf{F}_{\text{b,xy}} - 3\pi \eta D(v_b - v_f) + \mathbf{F}_{\text{mag}}$$  \hspace{1cm} (5)

where $\mathbf{F}_{\text{b,xy}} = -2\pi \eta D^3(v_b - v_f) \mathbf{z}$ is the unit vector in the $z$-direction, $g = 9.82$ m/s$^2$ is the magnitude of the gravitational

![Fig. 2. The assembled fluid system on the Peltier element-based temperature control system.](image-url)
acceleration, \( \rho_f = 1.0 \times 10^3 \; \text{kg/m}^3 \) is the density of water, \( \eta = 8.2 \times 10^{-4} \; \text{Pas} \) is the viscosity of water and \( \mathbf{v}_n \) and \( \mathbf{v}_f \) are the bead and fluid velocities, respectively. In this equation, we have neglected bead-bead interactions and interactions between the bead and the channel wall. Hence, it is assumed that the beads are ideal tracer particles for the flow.

First, we analytically calculate the sedimentation in the absence of magnetic forces. Second, we consider the magnetic force arising from the bias current. We will base all numbers on a bead mass-volume concentration of \( c = 0.1 \text{ kg/m}^3 \) corresponding to a bead number concentration of \( c_n = 9.7 \times 10^{13} \) beads/m\(^3\).

1) Bead Sedimentation Flux in the Absence of Magnetic Forces: By assumption, the only forces in the \( z \)-direction are the drag and buoyancy forces. It is found from (5) that

\[
v_z = \frac{D^2 (\rho_f - \rho_w) g}{18 \eta}.
\]

The bead velocity \( v_z \) along the channel is equal to the fluid flow velocity. We assume that the fluid velocity is only non-zero in the \( x \)-direction and only depends on \( y \) and \( z \), i.e., \( v_z = v_z(y, z) \). As no forces act in the \( y \)-direction, \( v_y = 0 \). Fig. 3 shows a bead trajectory and the definition of coordinates used in the calculations.

The number of beads entering the channel per unit time through an infinitesimal area \( \partial y_0 \partial z_0 \) in the \( yz \)-plane at \( (0, y_0, z_0) \) is

\[
\frac{\partial N}{\partial t} = v_z(y_0, z_0) c_n \partial y_0 \partial z_0.
\]

These beads land in the \( xy \)-plane on the infinitesimal area \( \partial x_1 \partial y_1 \) around \( (x_1, y_1, 0) \). Hence, the number of beads landing per area per time (the bead sedimentation flux) is

\[
\frac{\partial N}{\partial t \partial x_1 \partial y_1} = v_z(y_0, z_0) c_n \frac{\partial z_0}{\partial y_1} \frac{\partial z_0}{\partial x_1}.
\]

Since the velocity in the \( y \)-direction is zero, \( y_0 = y_1 \) and \( (\partial y_0/\partial y_1) = 1 \). To find \( \partial z_0/\partial x_1 \), we need the relation between \( \partial z_0 \) at the starting coordinates \( (0, y_0, z_0) \) and \( \partial x_1 \) at the landing coordinates \( (x_1, y_0, 0) \). The time \( t_0 \) it takes for a bead to sediment from an initial height \( z_0 \) over the channel bottom is \( t_0 = -z_0/v_z \). The distance traveled by the bead in the \( x \)-direction is

\[
x_1 = \int_0^{t_0} v_z(y_0, z(t)) dt = -v_z^{-1} \int_0^{z_0} v_z(y_0, z) dz.
\]

Hence

\[
\frac{\partial z_0}{\partial x_1} = \frac{\partial x_1}{\partial z_0} = \frac{v_z}{v_z(y_0, z_0)} = \frac{v_z}{v_z(y_0, 0)}
\]

which inserted in (8) results in the bead sedimentation flux

\[
\frac{\partial N}{\partial t \partial x_1 \partial y_1} = -v_z \cdot c_n.
\]

Thus, we have shown that the bead sedimentation flux is uniform and independent of the fluid flow velocity profile. Inserting the numbers for our MyOne bead suspension yields a bead sedimentation flux of \( 4.08 \times 10^{-7} \; \text{s}^{-1} \; \text{m}^{-2} \). For our sensor, which has an active area of \( 40 \times 40 \, \mu\text{m}^2 \), this corresponds to one bead landing on the sensor every 12.6 s.

To get a feel for the bead coverage, it is also convenient to express the bead sedimentation flux in terms of the number \( N_{ML} \) of monolayers of close-packed beads on the surface. Using that each bead takes up the area \((\sqrt{3}/2)d^2 \) and (11) and (6), we find

\[
N_{ML}(t) = \frac{\sqrt{3} \rho_f - \rho_w}{6\pi \mu_0} \frac{gD}{\eta} \cdot t_c.
\]

Inserting the values for MyOne beads we find \( v_z = -0.51 \; \mu\text{m/s} \) and that \( N_{ML} = 4.76 \times 10^{-6} \; \text{m}^3\text{kg}^{-1}\text{s}^{-1} \cdot \text{t} \). For a bead concentration of \( c = 0.1 \; \text{kg/m}^3 \) this means that a monolayer of beads should sediment in about 21.0 ks.

2) Magnetic Forces: In this section we consider the magnetic force on a bead arising from the bias current. The magnetic force is given by

\[
\mathbf{F}_{mag} \simeq \frac{1}{12} \pi D^3 \mu_0 \mathbf{H} \cdot \nabla \mathbf{H}^2.
\]

where \( \mu_0 \) is the permeability of free space and \( \mathbf{H} \) is the magnetic field intensity in the absence of the magnetic bead. This field has contributions due to the bias current passed through the sensor and the magnetostatic field from the ferromagnetic \( \text{Ni}_{81}\text{Fe}_{19} \) layer. In our study, we will restrict ourselves to study the former contribution. For an infinitely long surface conductor of width \( w = 40 \; \mu\text{m} \) placed along the \( x \) axis centered at \((x_c, 0, 0)\), the field at a point \((x, y, z)\) is found from Biot and Savart’s law as

\[
\mathbf{H} = \frac{I_x}{2\pi w} \left( \arctan \left( \frac{-4uz}{4z^2 + 4y^2 - w^2} \right) \mathbf{y} + \frac{1}{2} \ln \left( \frac{(y + w/2)^2 + z^2}{(y - w/2)^2 + z^2} \right) \mathbf{z} \right).
\]

When inserting this into (13), an analytical expression for the magnetic field is obtained. This expression is then evaluated in the space around the conductor slab. A contour plot of the \( z \)-component of the magnetic force divided by the buoyancy force is shown in Fig. 4 for the typical ac current amplitude of \( I_x,0 = 7.1 \; \text{mA} \) corresponding to a root mean square (rms) current of 5 mA. The magnetic force is found to be smaller than the buoyancy for all bead positions and is thus expected not to be important for the bead capture. As \( F_{mag,x} \) is proportional to \( I_x,0^2 \), contours for other currents can easily be found by adding \( 2\log_{10}(I_x,0/1 \; \text{mA}) \) to the contour values in Fig. 4. Even for an eight times higher current amplitude of \( I_x,0 = 56.8 \; \text{mA} \), \( F_{mag,x} \)
will only be larger than \( F_{\text{buoy}} \) up to 15 \( \mu \text{m} \) in the \( z \)-direction over the sensor edges. This distance is small compared to the channel height suggesting that gravitational sedimentation will dominate the accumulation of beads.

IV. RESULTS

Three measurement series were performed as a function of the bead concentration \( c \), the fluid flow rate \( Q \), and the bias current amplitude \( I_{x,0} \), where one parameter was varied, while the others were kept constant.

A. Bead Concentration Dependence

The bead concentration was varied between \( c = 0.025 \text{ kg/m}^3 \) and \( c = 0.2 \text{ kg/m}^3 \) for fixed \( Q = 50 \mu \text{L/min} \) and \( I_{x,0} = 7.1 \text{ mA} \). Fig. 5 shows measured values of the second harmonic out-of-phase response \( V''_{y,2} \) as a function of \( t \cdot c \), where \( t \) is the time after the bead suspension is injected into the channel. It is observed that the time traces of \( V''_{y,2} \) for different bead concentrations in

\[
\log_{10}(F_{\text{mag},z}/F_{\text{buoy}})
\]

Fig. 4. Contour plot of the \( z \)-component of the average magnetic force divided by the buoyancy force. The magnetic force is calculated for an ac current with amplitude \( I_{x,0} = 7.1 \text{ mA} \).

\[
V''_{y,2} \text{ [V]}
\]

Fig. 5. Values of \( V''_{y,2} \) measured as function of \( t \cdot c \) for the indicated bead concentrations \( c \). Fixed values of \( Q = 50 \mu \text{L/min} \) and \( I_{x,0} = 7.1 \text{ mA} \) were used in the experiments.

\[
Q = 10 \mu \text{L/min}
\]

Fig. 6. Values of \( V''_{y,2} \) measured as function of time, for \( Q = \{10, 20, 40, 160, \text{ and } 640\ \mu \text{L/min}\} \). Fixed values of \( c = 0.1 \text{ kg/m}^3 \) and \( I_{x,0} = 7.1 \text{ mA} \) were used in the experiments.

Fig. 5 fall onto the same curve. Moreover, the time traces of \( V''_{y,2} \) are linear.

For \( c = 0.025 \text{ kg/m}^3 \) and \( c = 0.075 \text{ kg/m}^3 \), the bead suspension flow was switched off after some time and an air bubble was injected followed by DI water to wash the beads out of the fluid system. In both cases, the signal is observed to return to its baseline level.

B. Flow Rate Dependence

The flow rate was varied between \( Q = 10 \mu \text{L/min} \) and \( Q = 640 \mu \text{L/min} \) for fixed \( c = 0.1 \text{ kg/m}^3 \) and \( I_{x,0} = 7.1 \text{ mA} \). Fig. 6 shows measured values of the second harmonic out-of-phase response \( V''_{y,2} \) as a function of \( Q \).

For small \( t > 0 \), the signal changes linearly with time as was also observed in Fig. 5. The initial slope, \( -\partial V''_{y,2}/\partial t \), varies systematically with \( Q \) and increases for increasing \( Q \). For longer times, the signals level off and approach their saturation values. The time at which this happens is shorter for high values of \( Q \).

Fig. 7 shows the time derivatives \( -\partial V''_{y,2}/\partial t \) of the initial response in Fig. 6 as function of \( Q \). The inset in Fig. 7 shows the same data on a log-log scale. It is seen that the data points are well described by a power law in the investigated interval. A fit yields the expression

\[
\frac{\partial V''_{y,2}}{\partial t} = 2Q^{\frac{1}{3}}(Q[\mu \text{L/min}])^{0.36}
\]

which is shown as the lines in figure.

In a preliminary study in [16] using the same fluidic system and bead type with \( c = 0.2 \text{ kg/m}^3 \) and a different sensor from the same wafer, the initial slope of the sensor response was found to have a linear dependence on \( Q \) within the significant experimental uncertainty. In the present study, the fluid handling was improved resulting in more reproducible experimental results.

C. Bias Current Dependence

To further investigate the capture of magnetic beads on the sensor, we studied the dependence of the measured sensor re-
and, as observed in Fig. 6, the signal was found to be a function of the time, where the signal levels off and approaches a saturation value, increases with increasing value of $Q$, and increase linearly with $c$ for constant $I_{x,0}$. Moreover, the absolute saturation value increases with increasing $I_{x,0}$. It is seen that the slopes of the normalized signals approximately assume the same value for all values of $I_{x,0}$. However, for each value of $I_{x,0}$, the time, where the signal levels off and approaches a saturation value, increases with increasing value of $I_{x,0}$. Moreover, the absolute saturation value increases with increasing $I_{x,0}$.

V. DISCUSSION

In the experiments where the bead concentration was varied for fixed values of $Q$ and $I_{x,0}$, the signal was found to be a function of $t \cdot c$ and increase linearly with $t$. Both of these results are in agreement with the predictions of the theoretical analysis of the gravitational bead sedimentation in (12). According to this analysis less than a single monolayer of beads should be sedimented on average on the surface for all experimental conditions. Thus, the observed linear change in the signal is consistent with a linearly increasing partial coverage of the sensor assuming that the sensor response is proportional to the bead coverage.

To discuss the signal dependence on $Q$, we first consider the effect of varying $Q$ on the gravitational sedimentation of beads. According to the theoretical calculations in (12), the influx of magnetic beads on the surface is constant. When it is taken into account that the fluid flow can also remove beads from the surface and that the probability of a bead being removed increases with increasing $Q$, one would expect zero or a negative change of $-\partial V_{y,2}^\prime/\partial t$ with increasing $Q$. Hence, the observations in Fig. 7 cannot be explained in terms of just gravitational sedimentation of the beads.

The magnetic force on a bead due to the applied current is strongly localized near the edges of the conductor (cfr. Section III-B2). The magnetic forces will be able to capture and hold beads passing close to the conductor even when $Q$ is large enough to tear sedimented beads away from areas in the channel with no magnetic forces. The contour defining the bead capture zone, i.e., the region where the magnetic force is strong enough to prevent beads from being torn loose by the flow, depends on both $I_{x,0}$ and $Q$.

The magnetostatic field from the Ni$_{81}$Fe$_{19}$ layer can also create capture zones as it gives rise to a force on the beads in the vicinity of the Ni$_{81}$Fe$_{19}$ layer edges with a nonzero normal magnetization component. That this effect plays a role can be observed in Fig. 9, which shows a micrograph of a dry sensor after exposure to the magnetic bead suspension and gentle washing. The beads are clearly observed to accumulate near the edges of the Ni$_{81}$Fe$_{19}$ layer parallel with the $y$ axis where there is a significant magnetostatic field due to the sensor magnetization in the $y$-direction.

The sensor signal from accumulating beads will initially be determined by the amount of beads entering capture zones on and near the sensor. Beads can either be captured from the flowing suspension by the short range magnetic forces or while rolling along the surface. Hence, $-\partial V_{y,2}^\prime/\partial t$ will grow with increasing $Q$, as observed in Fig. 6.
When sufficiently many layers of beads have accumulated in the capture zones, beads will begin to tear loose as the magnetic forces in the top layers are too weak to hold them. After a long exposure to the bead suspension, the amount and distribution of beads, thus, go towards an equilibrium defined by $Q$, $I_{x0}$ and the geometry of the Ni$_{81}$Fe$_{19}$ layer, as observed in Fig. 6.

The experiments performed for varying sensor bias current $I_{x0}$ showed that the initial values of $-\frac{\partial V_{av}}{\partial t}$ were approximately independent of the value of $I_{x0}$, but also that the saturation level varied significantly such that more beads accumulated on the sensor for high values of $I_{x0}$. This shows that the electromagnetic field from the bias current only plays a small role in attracting the beads to the sensor but is important for holding beads in the capture zones near the sensor. It also shows that the efficiency and size of these capture zones increase significantly with the current. These indications are in agreement with the theoretical considerations in Section III-B2, where it was shown that the magnetic force from the bias current is localized around the sensor edges.

VI. CONCLUSION

We have systematically investigated the capture of MyOne magnetic beads on magnetic sensors in a microfluidic channel under a fluid flow as function of the time, the bead concentration, the fluid flow rate and the sensor bias current. We have shown theoretically that the gravitational bead sedimentation flux is proportional to the bead concentration and sedimentation velocity and independent of the fluid flow profile. Experimentally, the sensor response was found to be a function of the time times the fluid flow rate and the sensor bias current. We have shown that the efficiency and size of these capture zones increase significantly with the current. These indications are in agreement with the theoretical considerations in Section III-B2, where it was shown that the magnetic force from the bias current is localized around the sensor edges.

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