Ultrasmall particle detection using a submicron Hall sensor

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We demonstrate detection of a single FePt nanoparticle of diameter 150 nm, moment $\sim 10^7 \mu_B$ using an ultrasensitive InSb Hall sensor with the bar lateral width of 600 nm. The white noise of a typical nanodevice, $S_V^{1/2} \approx 28 \text{ nV}/\sqrt{\text{Hz}}$, is limited only by two-terminal resistance of the voltage leads which results in a minimum field sensitivity of the device $B_{\text{min}} = 0.87 \text{ T}/\sqrt{\text{Hz}}$. To detect a single FePt bead, we employed a phase-sensitive method based on measuring the ac susceptibility change in a bead when exposed to a switched dc magnetic field. Such nano-Hall devices, enabling detection of potentially even smaller moments, are of considerable significance both for nanomagnetic metrology and high sensitivity biological and environmental detectors.

Properties of magnetic nanoparticles have been a subject of intensive theoretical and experimental studies since the late 1940s. Recently, various magnetic properties of individual particles and nanowires were successfully studied using a number of techniques, including GMR sensors, dc nano-SQUIDs –superconducting quantum interference device, and Hall magnetometry. All the methods listed above have their own advantages and vary in terms of magnetic sensitivity, applicability to particular magnetic systems/ experimental conditions and complexity. Small Hall sensors operating at room temperature are preferred for numerous biomedical applications. The main advantages of semiconductor Hall sensors are their noninvasive nature and high magnetic field sensitivity.^{1,2} Such sensors are already an essential part of many existing and prospective detection schemes for the biomedical industry, allowing a straightforward method for basic research into molecular recognition processes with potential single molecule resolution.

Among novel applications, it was proposed that a small submicron-sized Hall sensor could be an integrated part of a generic biosensor/nanoactuator device.³ Depending on the specific application, magnetic particles used as biological labels can be prepared in a variety of sizes and have an advantage of controllable tagging. For a number of applications –both related to biomedical and information technology –IT– industries – a precise knowledge of magnetic properties on the nanoscale is required. Despite enormous progress in the understanding of magnetic phenomena achieved so far, many fundamental questions are still unanswered. Among them are metrological issues related to measurements of ultrasmall magnetic moments. Thus, the magnetic investigation of a single nanoparticle is of special interest both from a fundamental/metrological point of view and for existing and potential applications, including magnetic recording, biomedical applications, and environmental monitoring.

Here, we present our results on detection of a small FePt nanoparticle with diameter of 150 nm and a magnetic moment of about $10^7 \mu_B$ using an ultrasensitive and noninvasive InSb Hall sensor. Although a number of attempts have been undertaken to detect small paramagnetic particles^{4–6} and even studies of the magnetization reversal in small ferromagnetic particles were reported previously,⁷ we present the first submicron-size Hall sensor capable of detecting an individual magnetic nanobead –composed of

5×10^4 nm size nanoparticles at room temperature. Undoped InSb films about 300 nm thick were grown by two-phase molecular-beam epitaxy on semi-insulating GaAs (001) substrates at a base pressure of 10^{-10} mbar. The growth procedure was adopted from Ref. 8. Initially a low-temperature ($T_{gr}=300$ °C) InSb passive buffer layer (approximately 20 nm) was deposited followed by the growth of an undoped InSb active layer ($T_{gr}=380$ °C). The method allows a significant improvement of structural and electronic properties of the InSb films. A high carrier (electron) mobility, $\mu = 1.3 \text{ m}^2/\text{Vs}$, and concentration, $n = 3.9 \times 10^{16} \text{ cm}^{-3}$, were deduced from magnetoresistance measurements in a perpendicular magnetic field in the van der Pauw geometry. Double Hall crosses with bar widths in the range of 600 nm–5 μm were fabricated by e-beam lithography and reactive ion etching (Fig. 1, left). The ohmic contacts were formed using a nonannealed titanium/gold layer on the electrical pads. As a result, a 600-nm sized Hall device has four-terminal and two-terminal resistances of 6.5 k Ω m and 58 k Ω m, respectively. The large two-terminal resistance is a result of long InSb leads. All experiments were performed at room temperature in a dark environment. The device was biased by a battery powered dc current source. As a part of the initial characterization, transverse or Hall (V_{xy}) and longitudinal (V_{xx}) voltages were measured on all of the devices. The typical value of the Hall coefficient is $R_H = 660 \text{ Ohm/T}$ (Fig. 2a) and magnetoresistance is $MR = 4.7\%$ for the as prepared devices with a smallest size of 600 nm. All adjacent Hall crosses demonstrate very similar characteristics with the difference in the Hall coefficient $\sim R_H/5\%$ where the smallest devices demonstrate the largest value, making these devices suitable for simultaneous measurements, when the magnetic particle is placed on cross no. 1, while cross no. 2 is left empty as a control experiment.

The Hall coefficient is weakly dependent on the size of the device, being $R_H = 660$ and 850 Ohm/T for crosses with bar widths of 600 nm and 5 μm , respectively. This decrease in Hall coefficient is associated with the stronger influence of interfaces and increased scattering of carriers in smaller devices, which lead to changes in mobility in the interfacial regions.

For noise measurements, the transverse voltage leads were directly connected to the input of a low frequency fast Fourier transform spectrum analyzer. All Hall devices

exhibit a $1/f$ like noise spectrum where the position of the $1/f$ -noise corner is proportional to the bias current and inversely proportional to the size of the Hall sensor (Fig. 2_b). Above this threshold, the noise is independent of frequency, bias current, and dc magnetic field, i.e., it is essentially white noise. For a typical 600-nm wide device the white noise is in order of $\sim 28 \text{ nV}/\sqrt{\text{Hz}}$ at $f \sim 70 \text{ Hz}$ and zero bias current (Fig. 2_b) and limited only by the relatively high two-terminal lead resistance of the Hall sensor. Thus, the minimum field sensitivity of the device achievable at high frequencies or low bias currents (white noise level) is defined as $B_{\text{min}} \sim 0.87 \text{ T}/\sqrt{\text{Hz}}$. However, at lower frequencies ($1/f$ noise) the spectral noise rapidly increases with the bias current and is inversely proportional to the bar width (Figs. 2_b and 2_c).

FePt beads were fabricated from the water-based solution of FePt nanoparticles. In the first step, FePt nanoparticles were synthesized in hexane by a high-temperature organometallic route using oleic acid and oleic amine as stabilizers.⁹ Then the particles were transferred into water using tetramethylammonium hydroxide as a phase transfer agent.¹⁰ The water-based nanoparticle dispersion was placed into an ultrasound bath at 80°C for 1.5 h that led to formation of beads $\sim 70\text{--}200 \text{ nm}$ in diameter composed of the FePt nanoparticles as shown in Fig. 1 inset. TEM analysis shows that the beads consist of a large number of closed packed FePt nanoparticles in an fcc chemically disordered state, with diameters of 3–4 nm and side-by-side distances of $\sim 0.5 \text{ nm}$. The energy dispersive x-ray spectroscopy reveals the 40:60 composition of a nanoparticle (Fe₄₀Pt₆₀).

Magnetization measurements were performed on a large ensemble of FePt beads and demonstrated their superparamagnetic properties at room temperature. The magnetic moment of the 150 nm sphere is $\sim 6 \times 10^7 \text{ B}$ in a field of $H \sim 30 \text{ kOe}$ as estimated by SQUID magnetometry and x-ray magnetic circular dichroism (XMCD) measurements.¹¹ A single FePt bead with a size of 150 nm was positioned on the top of one of the Hall sensors using a FEI focused ion beam (FIB) system equipped with a four-probe Zyvex nanomanipulation system. The second cross was left empty as a control experiment. A Si membrane fabricated in the FIB with a hole in its center was used to keep the bead in place and protect the InSb sensor from elevated temperatures and radiation during exposure to the electron beam (Fig. 1 right). As the membrane is $\sim 200 \text{ nm}$ thick and the

hole diameter ~ 120 nm is smaller than the bead, the estimated distance between the bead center and the Hall cross is ~ 225 nm.

For detection of a single FePt bead we have further advanced a phase-sensitive method initially described in Ref. [12]. The technique developed here is based on measurements of the nonlinear susceptibility change in a bead exposed to an ac magnetic field $B_{ac}=1.1$ mT, $f=491$ Hz applied perpendicular to the plane of the Hall sensor, as a function of an external dc magnetic field B_{dc} , applied parallel to the ac field. The Hall voltage is lock-in detected at the second harmonic $2f$ of the ac field frequency [Fig. 3]. Measurements at the second harmonic allow us to avoid the direct inductive pickup commonly occurring at the first harmonic [see e.g., Ref. 1]. While in the original detection scheme ac and dc fields are orthogonal, the amplitude of the $2f$ signal is proportional to the dc magnetization of the bead, in the method developed here ac and dc fields are parallel, the signal at $2f$ arises from the nonlinearity of the M_B curve and is proportional to the curvature of the M_B plot. A model, to be described in more detail elsewhere, has been developed which shows that the second harmonic signal rises from zero at $B_{dc}=0$ to a maximum at a value of B_{dc} corresponding to the maximum curvature of the magnetization curve, before falling toward zero as the magnetization approaches saturation.

The results of an experiment in which $B_{dc}=340$ mT was applied in a pulse mode are shown in Fig. 4, where the dc magnetic field was switched on/off three times, each pulse having a duration of 40 s. The cross with the magnetic bead shows a clear step-wise increase in the ac Hall voltage, $V_H \sim 150$ nV, with no detectable signal measured on the empty second cross. Our simulations show the expected step-wise second harmonic signal to be within a factor of 3 of this value, rather reasonable agreement since the signal is very sensitive to both the approach distance between bead center and Hall cross and the active dimensions of the nanoscale cross itself. It should be noted that the operational frequency range of the existing setup is limited to relatively low frequencies.

Limitations of the present detection scheme [i.e., parasitic inductance and capacitance signals at high frequencies] do not allow operation of the Hall devices at their white noise level [the crossover between $1/f$ and white noise occurs at ~ 5 kHz for the 600-nm

device at $I_H=50$ A where the sensitivity of the sensor would be maximal. Thus, further improvement of the measurement scheme will allow detection of significantly smaller magnetic moments.

In summary, we have present results on magnetic detection of a single 150 nm superparamagnetic FePt bead using a submicron-size InSb Hall sensor. Superior performance of the Hall devices studied here opens up new horizons for detection of even smaller magnetic moments, which is of high importance both for metrology of nanomagnetism and for highly sensitive biological and environmental detectors.

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FIG. 1. _Color online_ Left panel: An AFM image of the double-cross InSb Hall sensor with the bar width of 600 nm. A FePt bead _150 nm_ is highlighted for clarity _not to scale_. Inset: high-resolution transmission electron microscopy image of a bead. Right panel: A Si membrane with a hole in the center was used to keep the bead in place and protect the InSb sensor from elevated temperatures and radiation during the exposure to the electron beam. The FePt particle is located at the end of the manipulation tip just prior to placement in the hole.

FIG. 2. _a_ Hall coefficient measurements in two adjacent Hall crosses with size 600 nm. _b_ Spectral noise in the same device, $I_H = 0-50$ A. _c_ Current dependence of voltage noise in the devices with the lateral size 600 nm-5 μ m at $f=491$ Hz.

FIG. 3. Schematic of the detection experiment.

FIG. 4. Detection of an FePt bead, based on the ac-dc measurement technique. Arrows indicate switching of the dc magnetic field on and off, each pulse having a duration of 40 s. The cross with a magnetic bead shows a clear step-wise increase in the ac Hall voltage, V_{H_150} nV. No signal is measured on the empty cross no. 2.