Quantitative imaging of stray fields and magnetization distributions in hard magnetic element arrays

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In order to determine magnetic stray field and magnetization distributions of thin magnetic patterns and arrays, we developed a new quantitative imaging technique based on magneto-optical indicator films (MOIF) combined with inverse magnetostatic methods. The method is applied to hard magnetic FePt and PrCo$_5$ films which exhibit out-of-plane and in-plane easy magnetization axes, respectively. The films are patterned with standard electron beam lithography into square shaped elements with sizes between 10 $\mu$m and 500 nm. The magnetization values obtained from the MOIF method are compared to those of integral magnetometer measurements and show a good agreement, if the sensor properties are taken into account, properly. As an outlook, a concept for combining MOIF imaging with magnetic force microscopy is demonstrated which allows for quantitative magnetization imaging with a resolution down to 10 nm. © 2007 American Institute of Physics. [DOI: 10.1063/1.2717560]

I. INTRODUCTION

The growing research on magnetic patterns and nanostructures in recent years has increased the demand for advanced space-resolved magnetic imaging techniques.\(^1\) A main challenge is the ability to image the individual behavior of small structures in combination with the collective magnetic behavior and magnetic interactions of large patterned arrays. According to the physical mechanism of interaction between the probe and the sample, the different magnetic imaging methods usually either map the magnetization or the stray field distributions.\(^2\) The first concept is applied, e.g., in Kerr microscopy,\(^3,4\) different types of scanning electron microscopes equipped with a spin polarization detector\(^5,6\) and spin polarized scanning tunneling microscopy\(^7,8\) which are sensitive to the surface magnetization. X-ray magnetic circular dichroism (XMCD)\(^9,10\) is able to probe bulk magnetization. In contrast, different modifications of Lorentz microscopy [e.g., differential phase contrast (DPC),\(^11\) electron holography\(^12\)] and magnetic force microscopy (MFM)\(^13,14\) sense magnetic stray fields or stray field gradient distributions. For special sample configurations and geometries, inverse magnetostatic techniques then allow for a quantitative determination of magnetization components.\(^14-16\) Stray field sensitive techniques may be applied to a great variety of thin film samples and patterns, including electrically insulating materials and samples with degraded surfaces, where other surface sensitive methods fail.

An elegant method for the measurement of magnetic stray fields utilizes magneto-optical indicator films (MOIF),\(^17,18\) which are based on the use of the Faraday effect. The MOIF sensors can be calibrated with known external stray fields to achieve quantitative imaging. Being an optical technique, the spatial resolution of MOIF imaging is limited by the optical diffraction limit. Yet, the MOIF technique can be combined with another stray field imaging technique to achieve nanoscale spatial resolution. In fact, if the MOIF technique is used in combination with MFM, the spatial resolution of the combined method can be extended from the $\mu$m to the 10 nm range. Such a combined technique is particularly interesting for the nondestructive analysis of magnetic arrays,\(^19\) because it can provide information about magnetization and stray fields of both individual elements and large-element arrays. However, in general MFM is not a quantitative technique since the measured magnetic force depends on the MFM probe and the sample-probe interaction that is not a priori known. The most general ansatz for the determination of quantitative stray field values was put forward by Hug and co-workers\(^14,20\) based on a transfer function approach. Once the stray field is determined quantitatively by MFM, inverse magnetostatic techniques for the calculation of the magnetization are desirable.

In this paper, we focus onto the relation between stray field measurements and magnetization distributions of thin hard magnetic elements. We show that by application of the MOIF technique and inverse magnetostatic methods, quantitative stray field and magnetization imaging is possible on length scales between several millimeters to 500 nm. We solve the inverse problem for out-of-plane, as well as for in-plane, magnetization distributions in hard thin film magnetic elements. Particular emphasis is put on the effect of the...
finite thickness of the magneto-optically active layer (MOL) which leads to an averaging of the stray field perpendicular to the sensor plane (z direction) and contributes to a limitation of the lateral spatial resolution of the stray field measurement. The inverse magnetic problem is then considered in the limit of an effective measurement height approximation, where the z dependent stray field distribution \( H(x,y,z) \), measured within the finite sensor thickness, is replaced by an average \( H(x,y) \) at an effective height \( z_{\text{eff}} \). The inverse magnetostatic methods can also be applied to stray field distributions obtained by MFM or other techniques. Finally, a concept is outlined that allows for an advanced calibration of the MFM stray field signal. This concept is based on the inverse and forward magnetostatic techniques which we present in this paper.

II. EXPERIMENT

In order to demonstrate the potential of the combined MOIF and MFM method, permanent magnet films with well defined textures and easy magnetization axes perpendicular (L1\(_1\)-ordered FePt) and parallel to the film plane (PrCo\(_5\)) were patterned into arrays of square shaped elements by standard electron beam lithography (EBL). The FePt films were prepared by magnetron sputtering in an ultrahigh vacuum (UHV) via co-evaporation from elemental sources. The films with a thickness of 50–60 nm are grown on MgO (001) substrates using a substrate temperature of 650 °C. Before patterning, the orientation and chemical ordering of these films were controlled by x-ray diffraction measurements, indicating a c axis crystal structure with an ordering degree of about 85%. SQUID measurements [Fig. 1(a)] confirmed the perpendicular easy magnetization axis, revealing a coercivity of about 300 mT and a remanence of 92% of the saturation magnetization.

The PrCo\(_5\) films were grown epitaxially by UHV pulsed laser deposition (PLD) on Cr-buffered MgO (001) substrates at a temperature of 550 °C and covered with an additional 15 nm thin Cr-layer for oxidation protection. The crystallographic c axis, and thus the easy magnetization axis of the hexagonal PrCo\(_5\) crystallites has two possible orientations in the substrate plane, i.e., parallel to MgO [100] and MgO [010]. The texture in these films leads to strongly anisotropic magnetic properties [Fig. 1(b)]. The VSM measurement in the out-of-plane direction shows the typical hard axis loop behavior with a narrow and flat hysteresis. A measurement along one of the substrate edges probes combined hard and easy contributions due to the twofold easy axis configuration. This results in a large hysteresis with high coercivities of 1.5 T.

Arrays of square elements with various lateral sizes between 10 μm and 500 nm are written by EBL in a PMMA resist/developer system. The developed patterns are etched by 700 eV Ar-ions with a rate of 15 nm/min. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) reveal patterns with well-defined edges, where the sharpness of the pattern edges is limited by the natural edge roughness of the films of \( \approx 100 \) nm.

Magneto-optical imaging of the normal component of the magnetic field \( \mu_0 H_z \) above the surface of the magnetic elements is performed by using transparent ferrite garnet indicator films with a high Faraday rotation. The magneto-optically active layer (MOL) consists of a 600 nm thick Bi- and Lu-doped yttrium-iron-garnet film grown on a transparent gadolinium-gallium-garnet substrate. These films exhibit an in-plane easy magnetization and a specific Faraday rotation at visible wavelengths up to 4° μm\(^{-1}\). This maximum rotation occurs when the magnetization vector is tilted fully out-of-plane, i.e., along the z axis, which also is the direction of the light transmission. The saturation field is typically \( B_S = 40 \) mT and defines the possible range of field detection. The saturation field may be varied by the chemical additions and substrate orientations. An additional 30 nm thick Al mirror is deposited on the garnet film, in order to obtain high reflectivity and full double transmission of the polarized light across the MOL thickness. The choice of the mirror layer thickness and composition is one crucial step for a high sensitivity and spatial resolution. The basic principles of quantitative magnetic field imaging via MOIF films and the requirements for an advanced magneto-optical microscope are described extensively in Ref. 17. In order to improve spatial resolution, as well as magnetic sensitivity, a number of improvements have to be performed, including the stabilization of the optical system against vibrations, a reduction of the...
thickness of the MOL well below 1 μm and a minimization of the gap between the sample surface and the MOL.

The sensitivity of the MOIF technique to small stray fields in the range of 0.05–10 mT is strongly increased by applying an optical lock-in-technique, where the angular setting of the polarizer-analyzer system is modulated by a computer-controlled Faraday rotator. We use a 90° basic setting of the polarizer-analyzer angle α and a modulation angle of the Faraday rotator of δ≈±2°. Optimal field resolution with a linear calibration curve between measured photon counts and local magnetic field μ0H⊥ is obtained by differential detection of the α+δ and α−δ intensity distributions, acquired by a CCD camera with 1024×1280 pixels and applying a digital image processing system.

III. MAGNETOSTATIC PROBLEM

The magnetostatic problem is defined according to the Maxwell equations rot H=0 and div B=0, where the magnetic field in the exterior of the sample can be derived from a magnetostatic scalar potential H(r)=−∇φ via

\[ \phi(r) = \frac{1}{4\pi} \int \frac{\rho(r')}{|r-r'|} d^3 r'. \]

We consider an infinitely extended space with the boundary condition φ(r→∞)=0 and the magnetic charge density ρ(r)=div M(r) being localized in an area (x,y)∈A which is smaller than the spatial extend of the measurement plane. After the two-dimensional (2D) Fourier transform, Eq. (1) writes

\[ \phi(k,z > d/2) = \int_{-d/2}^{d/2} \rho(k,z') e^{i(k·z')} \frac{dz'}{2k}, \]

with k=(kx,ky), k=√k2x+k2y, and d denoting the film thickness of the magnetic structure. In order to invert this equation to determine a unique charge density from the measured field distribution, a z'-independent charge density has to be assumed. Consequently, we restrict our considerations to thin film geometries, where the z-dependence of M(r) can be disregarded or M(r) is approximated by the magnetization vector field averaged over the thickness of the film.

The two problems of the determination of in- and out-of-plane magnetization distributions must be treated separately, due to their different topological structure. In the case of a perpendicular magnetization, the location of the surface charge density is restricted to the z'=d/2 and z'=-d/2 planes. In contrary, for an in-plane magnetization distribution, ρ is an arbitrary function of the x and y coordinates, respectively. Consequently, the volume charge density is divided into two separate contributions

\[ \rho(k,z') = \sigma_\perp(k) \left[ \delta(z' - \frac{d}{2}) - \delta(z' + \frac{d}{2}) \right] + \rho_\parallel(k), \]

where the surface charge density \( \sigma_\perp(k,z) = M(k) \cdot n \) is related to the M⊥-component of the magnetization vector, with the normal vector n perpendicular to the surface. The volume charge density \( \rho_\parallel(k,z) \) is due to the in-plane magnetization components \( M_x \) und \( M_y \) with \( \text{div} \, M_r(k,z) = \rho_\parallel(k,z) \).

After performing the z’ integration, we obtain for the magnetostatic potential

\[ \phi(k,z) = \alpha(k,d,z) \left[ \sigma_\perp(k) + \frac{\rho_\parallel(k)}{k} \right], \]

with the transfer function

\[ \alpha(k,d,z) = \frac{e^{-zk}}{2k} \left( e^{kd/2} - e^{-kd/2} \right). \]

The z component of the magnetic stray field of the sample is then given by

\[ H_z(k,z) = -\frac{\partial}{\partial z} \phi(k,z) = k \cdot \alpha(k,d,z) \left[ \sigma_\perp(k) + \frac{\rho_\parallel(k)}{k} \right]. \]

In the following, the consideration is restricted to the two special cases of pure out-of-plane and pure in-plane magnetizations. For pure out-of-plane magnetization distributions the term in the square bracket of Eq. (6) reduces to \( \sigma_\perp(k) \).

The inverse problem to obtain the magnetization distribution from the measured stray field is solved by

\[ M_z(k) = \frac{H_z(k,z)}{k \cdot \alpha(k,d,z)}, \]

an equation which was already obtained in Ref. 14. For pure in-plane magnetization distributions, \( M_x(k)=M_y(k) \), the volume charge density can be determined via

\[ \rho_\parallel(k) = \frac{H_z(k,z)}{\alpha(k,d,z)}, \]

in Fourier space.

The determination of the in-plane magnetization components from \( \text{div} \, M_r = \rho_\parallel \) is only possible, if additional information or an additional equation (such as \( |M_r|=\text{const} \)) is applied. In the following, we consider hard magnetic materials, where we assume that the magnetization distribution of an element can be divided into mosaic-like patterns of piecewise constant magnetization vectors. They are separated by magnetic domain boundaries or the edge boundaries of patterned samples which have a width of \( 2\theta_\parallel \) below the measurement resolution. In this particular case, the magnetization distribution can be equivalently described by pure surface charges or by pure surface currents which are related to the jumps in the normal or tangential components of the magnetization, respectively. Both boundary value problems are equivalent, and, consequently, using such a tessellation ansatz, the total magnetization distribution can be obtained by the surface charge density, only. The in-plane volume charge density is then considered as a pattern of surface charge densities

\[ \sigma_\parallel(x,y) = \rho_\parallel(x,y) p, \]

where \( p \) denotes the lateral measurement resolution or the grid size of the discrete data. The in-plane magnetization may then be determined via...
\[ \sigma_i(x,y) = M_i(x,y) \cdot n_i, \]  
\[ n_i = \frac{\nabla \sigma_i}{|\sigma_i|}. \]

where the in-plane normal vector is then given by

In the following, we will summarize the validity and generality of the used ansatz. It is restricted to hard magnetic thin films and patterns, where the magnetization does not depend on the direction perpendicular to the film and the domain wall width may be disregarded. In this limit, which covers a lot of problems for patterned 2D systems, the obtained \( \mathbf{M(r)} \) are unique. A general problem for inverse problems is the instability of the solutions with respect to noise. The inverse transfer function \( \alpha^{-1} \) behaves as a high-wave vector amplifier and thus amplifies also all high-frequency noise components which are present in experimental data. The problem can be solved by filtering the high-frequency components, e.g., by means of a hanning window. Therefore, the signal to noise ratio and the measurement height \( z \) greatly influences the spatial resolution of the obtained magnetization patterns. In particular, for the in-plane magnetization reconstruction the noise can produce undesirable artifacts.

IV. EXPERIMENTAL RESULTS

Figure 2(a) shows the measured flux density distribution of the remanent state of an array of square shaped FePt elements after magnetization in an external field of \( \mu_0 H_{ex} = 1.3 \) T. All elements are fully magnetized and the stray field distribution above the pattern is measurable by our MOIF technique for all pattern sizes from 10 \( \mu \)m down to 1 \( \mu \)m. The resulting magnetization distribution according to Eq. (7) is shown in Fig. 2(b). For calculating the \( M_z(x,y) \) distribution, an effective measurement height for the MOL layer of \( z=200 \) nm was assumed which gives the correct remanent magnetization values of \( \mu_0 M = 1.3 \) T obtained from the superconducting quantum interference device (SQUID) magnetometer measurement of the same film before the patterning [Fig. 1(a)]. We will comment further on this topic in the discussion section of this paper.

Figure 3(a) shows the measured flux density distribution in \( z \) direction for an arrangement of four squares of PrCo\(_5\) elements. The image shows the remanent state of the pattern after applying an in-plane magnetic field of \( \mu_0 H_{ex} = 7 \) T in the \( y \) direction. The remanent magnetization is uniformly oriented in the \( y \) direction. Consequently, magnetic surface charges and a related magnetic stray field distribution are generated at the element edges parallel to the \( x \) axis only. Figure 3(b) displays the magnetization distribution of the marked \( 10 \times 10 \) \( \mu \)m\(^2\) square shaped element of Fig. 3(a). Due to the in-plane magnetization with surface charges only along two edges of the square, the spatial extend of the magnetic stray field in the \( z \) direction is much smaller compared to the case of perpendicular magnetization. Consequently, the magneto-optical response of the MOL is strongly inhomogeneous along the \( z \) direction and probably restricted to the lower part of the MOL. As can be seen in the magnetization distribution, the profile along the \( y \) axis is quite symmetric and constant, indicating that the surface charges at opposite points of the edge locations have nearly the same absolute value. The edge smoothing of the magnetization profile is a consequence of the limited spatial resolution. In the \( x \) direction the profile is more nonuniform and directly reflects a real inhomogeneity of the charge distribution along the edges. The stronger curvature cannot be explained by pure smoothing effects but is due to an inhomogeneous \( M_z \) distribution along the \( x \) direction. Most probable, this is caused by a local growth of domains with different easy magnetization directions along the \( y \) and \( x \) directions, respectively. Since the calibration function of the MOL assumes homogeneous Faraday rotation along \( z \), the obtained stray field values are underestimated and an effective measurement height of 400 nm has to be assumed in order to obtain good agreement with the remanent magnetization values of \( \mu_0 M_z = 0.6 \) T in the inverse problem. This remanence was determined by a VSM measurement of the nonpatterned film after saturation in a 9 T field [Fig. 1(b)].
A major difference in the application of the quantitative MOIF technique to small ferromagnetic patterns compared to large area flux imaging in superconductors is that the dependence of the magnetic stray field within the MOL thickness has to be taken into account in order to obtain a correct calibration of the MOIF signals. Figure 4 shows the measured flux density distribution for a 10 \times 10 \mu m^2 FePt square using a MOL with a thickness of 60 nm and a thickness of the Al mirror \(d_0=30\) nm. The MOIF sensor was pressed on top of the magnetic structures. Consequently, the MOL measures the perpendicular component of the stray field at a distance between 30 and 630 nm above the upper surface of the magnetic film. Figure 4 shows the calculated stray field distribution for the same geometry and a remanent magnetization of \(M_r=1.3\) T which was obtained by measuring the same film in a SQUID magnetometer. Assuming that the magnetization of the MOL directly follows the field lines of the FePt stray field, the \(H_z(z)\) values have been averaged over the measurement distance according to

\[
\bar{H}_z(x,y) = \frac{1}{d_{\text{MOL}}} \int_{d_0}^{d_0+d_{\text{MOL}}} H_z(x,y,z')dz'.
\]

Furthermore, in order to take into account the finite measurement resolution, the calculated \(H_z(x,y)\) distribution was slightly smoothed by applying a Gaussian filter with a width of \(\Delta x=0.8\) \mu m corresponding to an overall resolution limit of the combined sensor and optical system. This procedure yields a very good agreement between measurement and calculation. These results show that the \(H_z(x,y)\) signal measured with the MOIF technique represents to a high degree a linear superposition of the \(H_z(x,y,z)\) values for \(d_0\leq z\leq d_0+d_{\text{MOL}}\).

Based on these considerations, a second important step is the determination of an effective measurement height for the solution of the inverse problem. The ideal condition that the measurement of \(H_z(x,y,z)\) is performed with a probe of zero extension in the \(z\) direction can be neither fulfilled by
MOIF nor by MFM. Therefore, a wave number dependent sensor or tip transfer function is required which takes into account such properties as the finite thickness of the sensor. Yet, such an approach is beyond the scope of this paper. Instead, based on the above results that the measured \( H_0(x,y) \) signal represents a linear superposition of the real \( H_0(x,y,z) \) values, we approximate the field distribution for the inverse problem by using \( H_0(x,y) \) at an effective measurement height \( z_{\text{eff}} \). \( z_{\text{eff}} \) is the height above the sample, where the stray field \( H_0(x,y) \) equals the averaged stray field. This is determined once for a MOIF sensor by a comparison of the experimental \( H_0(x,y) \) distribution with a simulation of the forward problem. The simulations showed that for pattern sizes \( W \) larger than five times the resolution limit \( \Delta x = 800 \, \text{nm} \), e.g., for squares with 10 \( \mu \text{m} \) edge length, the decrease of \( \mu_0 H_0 \) with increasing measurement height \( d_0 \leq z \leq d_0 + d_{\text{MOIF}} \) is very weak: a variation of, e.g., \( \pm 250 \, \text{nm} \) in \( z_{\text{eff}} \) leads to a difference of only \( \pm 7\% \) for the magnetization value.

In contrast, for the quantitative magnetization measurement of structures with smaller \( W \) an increasing error of the \( M(x,y) \) distribution has to be taken into account. The reason for this effect is mainly that the field of smaller structures decays on smaller length scales along \( z \). A full quantitative treatment of this effect in the framework of a wave-vector-dependent transfer function is beyond the scope of this article. As revealed by stray field calculations of magnetic patterns with different \( W \), the limit of the effective height approximation is still valid to some extend for \( W \geq 2 \Delta x \). For smaller \( W \), for example, for a 1 \( \mu \text{m} \) FePt square with a homogeneous magnetization of 1.3 \( T \) \( (W = 1.25 \Delta x) \), a deviation of \( z_{\text{eff}} = \pm 100 \, \text{nm} \) results in a difference of \( \mu_0 M = \pm 0.2 \, T \) or \( \pm 15\% \). The effect of \( z_{\text{eff}} \) for different \( W \) can be seen in Fig. 2(b), where slightly lower magnetization values are determined for the 5 \( \mu \text{m} \) squares compared to the 10 \( \mu \text{m} \) ones. Summarizing, once calibrated, the effective height can be applied to all further quantitative measurements for \( W \geq \Delta x \) if the orientation of the magnetization is kept fixed. The latter restriction results from the fact that the stray field distributions of the strongly localized \( \sigma_{x,y} \) distributions exhibit a short range decay along the \( z \) direction and are additionally smoothed by the limited lateral resolution. As a consequence, different \( z_{\text{eff}} \) values have to be applied for the perpendicular and in-plane magnetizations.

One further goal of this work is to combine the MOIF measurements with MFM imaging, in order to increase the spatial resolution. Based on MOIF measurements and inverse and forward magnetostatic techniques a solid basis for the calibration of the MFM signal can be provided. MFM measurements can be performed in a noncontact linear lift mode where the cantilever scans the surface line by line at a constant lift height \( h \) of the tip apex. The highest sensitivity can be obtained in the dynamic force microscopy mode, where the MFM cantilever is forced to oscillate. The position dependent frequency shift \( \Delta f \) or the phase shift \( \Delta \phi \) of the forced oscillation is mapped. Figure 5(a) shows a MFM image for the same state of the FePt element array as in Fig. 2. The image was acquired at a measurement height of \( h = 100 \, \text{nm} \) with a commercial perpendicularly magnetized nanosensor tip. It can be shown that in 2D Fourier space the MFM phase contrast is related to the stray field gradients by a tip dependent transfer function ICF \(^{14} \)

\[
\Delta \Phi = \text{ICF}(k) \cdot \frac{\partial H_z(x,y)}{\partial z},
\]

where \( k \) represents the two-dimensional wave-vector in the \((x,y)\) plane. In extending the work of Hug et al., where an assumption about the magnetization distribution inside the sample is necessary, the local quantitative determination of \( M(x,y) \) by MOIF and inverse magnetostatic methods will give a more general basis for tip calibration. The \( \partial H_z(x,y,z)/\partial z \) distribution at the lift height of the MFM tip may be directly calculated, based on the determined \( M_z(x,y) \) distribution [see Fig. 5(b)]. A comparison between the \( \partial H_z/\partial z \) distribution and the \( \Delta \Phi \) distribution in the 2D \( k \) space can then be used to determine the transfer function ICF, i.e., for a calibration of the MFM tip. If, as a first approximation the \( k \) dependence of the transfer function is disregarded, the MFM image in real space then should be proportional to the field gradient distribution. It can be seen in Fig. 5, that there is a very good qualitative agreement between both images. The remaining difference can be attributed to the instrument transfer function. More details on sensor and instrument transfer functions for full quantitative
MOIF and MFM calibration are out of the scope of this paper and will be published in a forthcoming article.

VI. SUMMARY

We have demonstrated the potential of an advanced magneto-optical setup based on very sensitive MOIF films for full quantitative measurements of the normal component of the stray fields above the surface of thin magnetic elements. Our general inversion scheme allows for the quantitative determination of the in- and out-of-plane magnetization components, after the effective measurement height of a used MOIF sensor is determined. An excellent agreement between locally determined $M(x,y)$ values and global magnetization measurements is obtained. Since the MOIF sensors are slightly pressed on top of the magnetic structures by adjustable contact fingers, the measurement height can be reproducibly achieved and can be, therefore, considered as a property of the sensor for a given magnetization orientation and resolution. The solution of the problem of differences of $z_{eff}$ for in-plane and out-of-plane magnetization distributions requires a strong reduction of the MOL thickness in future or a more advanced description of the sensor by a sensor transfer function. After having determined the magnetization distribution, the magnetic field and gradient field distributions can be calculated at different measurement heights. The quantitative MOIF technique combined with inverse and forward magnetostatic techniques has the potential to provide a solid basis for the calibration of MFM signals. The essence of the concept was demonstrated for the simple example of a patterned FePt film with perpendicular magnetization. The potential of this calibration method is founded on the possibility of using a broad range of $k$ vectors for determining the tip transfer function. In our future work, a reliable calibration of the MFM will be established and then be used for developing a fully quantitative combined MOIF and MFM technique.