The magneto-optical gradient effect in an exchange-biased thin film: experimental evidence for classical diffraction theory

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Abstract. The magneto-optical gradient effect decorates the boundaries of in-plane domains even at perpendicular incidence of light in an optical polarization microscope. For its explanation, the classical magneto-optical diffraction theory was previously used to derive the effect from the same gyrotropic interaction as the Kerr effect. In order to explain the symmetry of the experimentally observed contrast on bulk ferromagnetic crystals, planar as well as perpendicular subsurface gradients in the magnetization had to be assumed. This was particularly needed when the surface magnetizations in neighboring domains pointed head-on and a gradient contrast appeared also in conditions of vanishing gyrotropic interaction at the surface. The gradient contrast in such conditions should not appear in very thin films where perpendicular magnetization gradients are not enforced by reduction of magnetostatic energy. Here we present the first experimental confirmation of this expectation, thus closing an experimental gap in verifying the predictions of the diffraction theory.

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1. Introduction to the gradient effect: discovery and phenomenology

After the introduction of image processing to magneto-optical Kerr microscopy in the mid-1980s of the last century [1, 2], the method has rapidly evolved into a powerful domain imaging technique, allowing us to visualize the surface domains of almost all kinds of magnetic materials at a maximum lateral resolution of roughly 300 nm. For a review of the possibilities of digitally enhanced Kerr microscopy, we refer to the monograph Magnetic Domains [3], in which most presented domain patterns have been imaged by this technique. The imaging capability also comprises the surface magnetization (Néel-cap) of vortex domain walls in low-anisotropy bulk materials, even if the wall width is below resolution like in the case of bulk iron. (A vortex wall is observed in bulk material and is related to the asymmetric Bloch wall in thick films. It is characterized by a Bloch-like magnetization rotation in the volume and a Néel-like rotation at the surface [3].) A diffraction-broadened image is then obtained [4], which nevertheless reveals details of the wall, such as internal Bloch lines (indirectly seen at the surface by characteristic surface kinks [5]) or wall segments of opposite surface rotation sense. There is just one micromagnetic object that can be hardly detected by magneto-optical means: at the transition region between two Néel caps of opposite rotation sense, cross or circular vortices with in-plane magnetization rotation are formed for topological reasons. In the center of a vortex a magnetization swirl [3] is located, which is magnetized perpendicular to the surface with a diameter of roughly 10 nm in the case of iron.

The attempt to image magnetization swirls by Kerr microscopy was the starting point of systematic magneto-optical domain wall studies that we (RS together with A Hubert) performed about 20 years ago on iron-silicon sheets with well-oriented (100)- and (110)-surfaces. To possibly see the swirls in wall transitions, perpendicular incidence of light has to be chosen. As the Kerr rotation is proportional to the sample magnetization component parallel to the propagation direction of the reflected light beam, all in-plane magnetized structures should not lead to a Kerr contrast under this condition. This applies to the Néel caps of the domain walls and to the surface domains, whereas a polar Kerr effect is to be expected at the locations of the swirls with their out-of-plane magnetization. At that time, we could not identify swirl contrast—swirls have been imaged later by magnetic force [6] and spin-polarized tunneling microscopy [7].

What we found instead [8, 9] was an unexpected contrast at all types of 180° and 90° domain walls present in the investigated samples, which could not be explained by any of the known magneto-optical effects. The contrast appeared in the Kerr microscope when the analyzer was nearly crossed relative to the polarizer and when a phase-shifter (rotatable λ/10-compensator), placed right in front of the analyser in the reflected light path, was ‘opened’ by some degrees. At certain orientation angles, the walls then showed up in a homogeneous black
Figure 1. Symmetry of the magneto-optical domain boundary contrast due to the gradient effect for in-plane magnetization, perpendicular illumination and horizontal polarization for three domain geometries: (a, b) 180° walls, (c, d) head-on boundaries that represent V-lines of an underlying flux-closure structure in bulk material and (e, f) head-on walls in thin films. For each pair of images, the contrast before and after a sample rotation by 90° is shown. Cases (a–d) have been found experimentally [8], whereas cases (e, f) have not been verified so far (after [10]).

or white color, which could be quantified by integrating line-scans across the digitized wall images. The contrast is independent of the rotation sense of the wall magnetization (which can be sensed by the Kerr effect at oblique incidence), and it does not depend on the wall width as could be proven by measurements on a variety of materials [9]. Therefore, it is better called ‘boundary’ contrast rather than wall contrast. Figures 1(a) and (b) schematically demonstrate the contrast symmetry for the case of regular 180° walls. Neighboring domain walls appear in alternating black and white, and the boundary contrast is inverted when the sample is rotated by 90°. It can also be inverted by rotating the compensator through its extinction position (different from the Kerr contrast, which usually is generated and inverted by the opening and rotation, respectively, of the analyzer alone). This indicates a phase shift of the magneto-optical amplitude of the boundary contrast of roughly 90° relative to the phase of the Kerr amplitude. Interestingly, also V-lines (figure 1(c) and (d)) obey the same contrast symmetry. V-lines are the surface intersections of internal domain walls that are arranged in the shape of the letter V (see figure 1, inset). Around a V-line the \( m \)-vectors of the surface domains are head- or foot-on, but underneath the surface the magnetization flow points away from (or toward) the surface. In this way, the magnetic flux of the surface domains is distributed in a stray-field-free manner—a stray-field-free magnetization field (\( \text{div} \ m \approx 0 \)) is naturally enforced in low-anisotropy materials, including iron [3].

A closer inspection of the contrast symmetry reveals that the boundary contrast is primarily determined by the magnetization jump from one domain to the other, i.e. by the magnetization gradient across the wall, and not by the properties of the actual wall. In analogy to the
Figure 2. Symmetry of the gradient contrast at neighboring 180° domain walls, derived from the dielectric law (1). The sample is rotated by 90° from (a) to (c). The positive and negative $D_y$ components are aligned along the analyzer axis, but at opposite directions (d). Parallel to $D_y$ are the magneto-optically reflected $E_y$-components. Due to their phase shift relative to the ordinary reflected $E_x$, a black/white microscopic contrast is created by matching the phase of one of the two $E_y$ vectors with that of $E_x$ by means of the compensator. The domain boundary effect at 90° walls and V-lines can be derived in the same way.

magneto-optical Voigt effect—a birefringence effect that depends quadratically on the magnetization vector and that shows a similar symmetry\(^6\)—also the domain boundary effect can be interpreted as a birefringence effect. It depends linearly on certain components of the gradient of the magnetization vector $m$ and may therefore be interpreted as ‘gradient effect’. In \([8, 12]\), a dielectric tensor was suggested to phenomenologically describe the high symmetry of the observed boundary contrasts. The $y$-component of the displacement vector $D$, which represents the magneto-optical amplitude of the gradient effect at perpendicular incidence and polarization along the $x$-axis, is written as

$$D_y = P \left( \frac{\partial m_x}{\partial x} - \frac{\partial m_y}{\partial y} \right) E_x,$$

where $x$ and $y$ are the coordinates along the $E$- and $B$-vectors of the incident light wave, respectively, and $P$ is a material constant that scales with that of the Kerr effect. A graphical representation of this law is presented in figure 2 for the case of two neighboring 180° domain walls. Relation (1) describes all 180°, 90° and V-line boundaries found on bulk material.

2. Physical explanation of the gradient effect

Although the symmetry of the domain boundary observations in \([8]\) could be well reproduced by the empirical dielectric law (1), the question of the physical origin of the effect remained open. Originally, it was thought that a derivation of the gradient effect from the gyrotropic

\(^6\) The Voigt or Cotton–Mouton effect is well known for domain decoration in transmission experiments. In connection with our wall studies, we have discovered this effect in reflection \([8]\) (an analogous reflection effect was very recently also seen in x-ray spectroscopy \([11]\)). The Voigt effect cannot explain the domain boundary contrast \([8]\), although both effects occur under the same experimental conditions and follow the same rotational symmetry.

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interaction of the Kerr or Faraday effect \( D \propto m \times E \) was not sufficient. Consider the three cases of figures 1(a)–(c): for a given horizontal polarization direction of light, a non-vanishing cross-product would indeed exist for all the three cases. The incident light, which is polarized along the \( x \)-axis, thus generates in each domain an oscillating gyrotropic polarization normal to the sample surface, the amplitude of which depends on the magnetization direction of the illuminated domain. The active magnetization component varies between zero (if \( m \) is along the \( x \)-axis) and 1 (if \( m \) is along the \( y \)-axis). In the neighboring domain, a similar oscillation is generated that is out of phase to the first one by \( 180^\circ \) since the \( m_y \)-component, which causes the gyrotropic interaction, changes sign (i.e. has a gradient across the boundary). The pairs of out-of-phase oscillating electric dipoles produce a quadrupolar field with a component perpendicular to the domain boundary and proportional to the gradient of \( m_y \). In the experiment, the component of this transverse field parallel to the analyzer axis (\( y \)-axis) can be detected. This detectable \( E_y \)-component is obviously proportional to the component \( \partial m_y / \partial y \) of the magnetization gradient and may be expressed as

\[
E_y^{(x,y)}(y, 0) \sim -B(\partial m_y / \partial y) E_0^x,
\]

with some constant \( B \). Although this measured component includes only the second term in (1), the boundary contrast of the three cases in figures 1(a)–(c) and the rotational symmetry between figures 1(a) and (b) (compare figure 2) can be explained with the term (2) alone, on the basis of gyrotropic interaction. This fact was recognized soon after the discovery of the gradient effect, and the boundary contrast of the mentioned cases was derived [13] by formal diffraction analysis under the assumption of only in-plane gradients.

This concept, however, does not work for the V-line orientation in figure 1(d): here the gyrotropic interaction between the horizontally oriented \( E \)-vector of the light and the in-plane domain magnetization \( m \) vanishes identically—the cross-product \( m \times E \) is zero, thus, it cannot give rise to any contrast even by diffraction. (A significant contribution to the observed integrated intensity of the gradient lines, emerging from the rotating wall magnetization itself, can be excluded [9].) In order to include also the contrast symmetry of the V-lines in the initial phenomenological description [8], the first term \( (\partial m_x / \partial x) \) in (1) had to be added empirically. But what is the origin of this term?

The interpretation of this (necessary) term requires consideration of polar gradients \((\partial m_z / \partial z)\), as was discovered in successive articles on the diffraction analysis [14]–[16]. The crucial point is the micromagnetics of a V-line: around a V-line the \( m \)-vectors are arranged in a stray-field-free pattern \((\text{div} \ m \approx 0)\). As there is an in-plane contribution to the divergence \( \partial m_x / \partial x \), it must be compensated by an almost equal but opposite \( \partial m_z / \partial z \) to fulfill the condition \( \text{div} \ m \approx 0 \). Even if the \( m_z \)-component is zero at the surface (all attempts to observe such a perpendicular component at V-lines have failed so far), it must rise below the surface and give rise to a non-uniform polar Kerr effect according to its limited but significant information depth of about 20 nm in metals [17]. In fact, by considering both, diffraction due to planar and perpendicular gradients in the analysis [15], it was possible to derive the empirically observed symmetry of the gradient effect including the critical case of figure 1(d) if the condition \( \text{div} \ m = 0 \) is valid.

This can be verified by extending our previous analysis: polar magnetization components cause a gyrotropic interaction for any wall orientation, so that the \( y \)-component of the electric field can be written as

\[
E_y^{(z)} \sim -\frac{1}{2} B(\partial m_z / \partial z) E_0^x.
\]

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The proportionality of the two constants in (2) and (3) follows from the symmetry of the magneto-optic interaction considered in the diffraction theory [16]. In bulk low-anisotropy materials, the flux-closing condition \( \text{div } \mathbf{m} \approx 0 \) implies
\[
\frac{\partial m_x}{\partial x} + \frac{\partial m_y}{\partial y} + \frac{\partial m_z}{\partial z} = 0,
\]
so that in this case, (3) may be replaced by
\[
E_{y}^{(c)} \sim \frac{1}{2} B (\frac{\partial m_x}{\partial x} + \frac{\partial m_y}{\partial y}) E_{x}^{0}.
\]

The full detectable \( y \)-component then is the sum of (2) and (5):
\[
E_{y}^{(x,y,z)} \sim \frac{1}{2} B (\frac{\partial m_x}{\partial x} - \frac{\partial m_y}{\partial y}) E_{x}^{0},
\]
which is consistent with the empirically found law (1). So under the condition \( \text{div } \mathbf{m} \approx 0 \), the gradient contrast can indeed be described by (1). A more systematic analysis of domain diffraction phenomena, including the gradient effect, was presented in a later article [10] that has completed a series of papers [17]–[20] dealing with magneto-optical interference and diffraction effects in magnetic multilayers. The pioneering paper [17] of this series, in which the concept of magneto-optical depth sensitivity was introduced, also contains a descriptive explanation of the subsurface gradient contribution based on the depth sensitivity function.

The results of the diffraction analysis can be summarized in several essential points. The gradient effect can, like the Kerr effect, be derived from gyrotropic interaction, but its symmetry is strongly affected by the micromagnetic conditions in the underlying domain structure. To explain the high symmetry of the effect, which was found experimentally and which is phenomenologically described by the dielectric law (1), two different contributions to the magneto-optical amplitude have to be considered: an effect of diffraction due to planar gradients and an additional effect due to perpendicular subsurface gradients. These two effects are highly correlated under the condition \( \text{div } \mathbf{m} = 0 \), i.e. for charge-free boundary configurations. In these cases, the diffraction analysis can even explain the gradient contrast of domain geometries in which a gyrotropic interaction is absent in the surface layer (like in the head-on domains in bulk material shown in figure 1(d)). For such domain boundaries, the polar subsurface magnetization gradient provides the additional gyrotropy required for the combined gradient effect. The symmetry of the gradient effect of charge-free configurations can be phenomenologically described by the dielectric law (1), which explicitly contains only in-plane magnetization gradients, while the perpendicular gradients that may be present in order to fulfill the condition \( \text{div } \mathbf{m} = 0 \) are taken into account implicitly in this condition.

The situation is different and simpler in thin films magnetized strictly in-plane, without the interfering contributions of perpendicular magnetization. In accordance with the above derivation of (2), a boundary between domains magnetized head-on as in figures 1(e) and (f) should give a strong gradient contrast when their magnetizations are perpendicular to the polarizer axis (maximum gyrotropic interaction, figure 1(e)), but no domain boundary contrast when both domain magnetizations are parallel to the polarizer axis (figure 1(f)). Such strict head-on configurations are not easily realized because they are connected with high magnetostatic energy. Equation (2) implies that the empirical relation (1) should be replaced by the simpler relation
\[
D_y \sim -(\frac{\partial m_y}{\partial y}) E_x,
\]
Figure 3. (a) Magnetic structure of the bimodal exchange-bias film used for the magneto-optical studies. The exchange bias field $H_{eb}$ changes sign in a stripe arrangement. This causes head-on domains in the NiFe film (b). The stripe width (domain width) was 20 µm in our sample.

3. Supportive experiment for diffraction theory

So far, there has been no experimental evidence for this prediction, which would support the adequacy of the diffraction theory. At the time when the contrast symmetry of figures 1(e) and (f) was predicted [10], straight domain boundaries surrounded by strictly head-on magnetized domains were actually not known in magnetic films. If head-on domains are enforced in thin films (e.g. by a gradient magnetic field or by magnetoelastic effects), they are separated by metastable domain boundaries that are zigzag folded to reduce magnetic charges [3, 22]. In the neighborhood of a zigzag wall, the magnetization tends to be oriented along the wall to further lower the charge density (see figures 3.83 and 5.61 in [3]). Furthermore, for an experimental proof of the gradient effect, at least three head-on walls should be arranged in close neighborhood to unambiguously see the alternating boundary contrast. Such configurations can hardly be produced in standard films due to the metastable character of these walls. Continuous longitudinal recording media would offer this geometry, but here again the bit transitions are zigzag folded for the mentioned reason and, in addition, the media are usually too rough to allow reasonable magneto-optical microscopy.

In ion-irradiated exchange-bias film systems [23], however, it is possible to generate the required domain and wall configuration. The sample basically consists of an exchange-coupled IrMn (antiferromagnet) and NiFe (ferromagnet) bilayer film. An exchange anisotropy direction is defined by the application of a magnetic field during film deposition. Subsequent local ion irradiation of the stack in the presence of an inverted magnetic field locally inverts the direction of the exchange anisotropy [23, 24]. A stripe pattern with antiparallel exchange bias $H_{eb}$ and the desired head-on domains in the NiFe film can be generated thereby as illustrated in figure 3. The domains are separated by straight domain walls. Owing to the NiFe film thickness of 20 nm, these are most likely of Néel character with an in-plane rotation of magnetization. Although the NiFe film, in which the domains and walls are imaged, is covered by the IrMn and a protection layer, its magneto-optical contrast is still sufficiently strong for domain imaging.
Figure 4. Magneto-optical microscopy on the IrMn/NiFe stripe structure. (a) The unprocessed contrast, in which the implanted and non-implanted areas with opposite exchange bias are revealed by different colors. (b)–(f) Difference images with the saturated state as a reference image. They show pure magneto-optical contrast, obtained at oblique illumination in the longitudinal Kerr effect (the plane of incidence is indicated by double lines) and at perpendicular illumination (marked by circles) under conditions of the gradient effect. Note that in the case of the longitudinal Kerr effect the plane of incidence coincides with the sensitivity direction. The polarization direction is horizontal in all images.

On this film system, we could indeed verify the predicted contrast symmetry as shown in figure 4. Here, a combination of images, obtained in the longitudinal Kerr effect at oblique incidence of light and under conditions of the gradient effect, i.e. perpendicular incidence, is presented. The polarization direction of the illuminating light is horizontal in all images. Head-on domains, which are magnetized transversely to the polarization axis, show an alternating black and white gradient contrast for neighboring domain walls (figure 4(c)) if imaged at perpendicular illumination. This domain boundary contrast is—as expected for the gradient effect [8]—independent of the true wall magnetization that is revealed by longitudinal Kerr microscopy in figure 4(b) (figures 4(b) and (c) are images of the same configuration). Together with the gyrotropic interaction, the gradient contrast disappears after rotating the sample by 90° (figure 4(f)). A residual double contrast is then left at each wall; it is caused by the superposition of Voigt contrast and local gradient contrast due to the wall rotation [9]. The wall magnetization of the state in figure 4(f) is shown in figure 4(e) together with the domain contrast in figure 4(d). The images in figures 4(c) and (f) prove the prediction illustrated in figures 1(e) and (f).
4. Conclusion

In conclusion, we can indeed verify that the contrast symmetry of the magneto-optical gradient effect in magnetic films follows the prediction [10] for diffraction on in-plane magnetization gradients. Our observation thus closes an experimental gap and adds further support to the adequacy of the diffraction theory of the gradient effect. It can (like the Kerr effect) be fully explained by gyrotropic interaction, and the phenomenological additions to the dielectric law [8, 12] seem not to be necessarily considered as new intrinsic contributions.

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