Magneto-optical additivity in ferromagnetic bilayers separated by nonferromagnetic spacers

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(Received 21 February 2005; accepted 14 June 2005; published online 8 August 2005)

We show that for some ultrathin multilayer structures the proportionality of the longitudinal Kerr ellipticity signal to the magnetic film thickness does not take place. Instead, thinner magnetic layers give even twice larger contributions than the thicker ones located a few monolayers deeper. This effect is demonstrated both experimentally and theoretically on ultrathin Fe/Cr/Fe sandwich systems prepared on GaAs(001) surfaces. Such a behavior is explained by a depth variation of phase relations between the optically reflected wave and the wave generated by the magneto-optical interaction. This mechanism becomes very effective close to the phase where the magneto-optical ellipticity cancels. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1996827]

I. INTRODUCTION

The magneto-optical (MO) Kerr effect is a convenient technique for studies of magnetic properties of ultrathin films and multilayers because of its high sensitivity and relatively low requirements on the experimental setup. This is true even when performing measurements in ultrahigh-vacuum (UHV) conditions and/or at low temperatures.

Starting from the electromagnetic theory a quite simple rule was derived for the polar (i.e., with the magnetization vector perpendicular to the film plane) and longitudinal (i.e., with the magnetization vector in the film plane and in the plane of incidence) MO geometries.[1] The simplification is based on the fact that the magnetic films or even quite complicated multilayer structures prepared on an absorbing substrate can be considered ultrathin. This simply means that the optical wave during propagation through the multilayer stack does not change considerably its amplitude and its phase. The mathematically described rule can be summarized as follows. (a) The magneto-optical observables (i.e., the MO Kerr rotation and the MO Kerr ellipticity) are sums of contributions originating in all magnetic layers involved. (b) Each of these individual contributions can be evaluated as if the given ferromagnetic layer was appearing on the substrate alone. This means that the optical effect of the other ferromagnetic or nonferromagnetic layers can be neglected. (c) The contribution of each ferromagnetic layer is proportional to its thickness. This expectation was confirmed experimentally for Fe(110)/Ag(111) multilayers,[2] for Co overlayers on Cu substrates, and for Co/Cu superlattices[3] studied *in situ* under UHV conditions. In both cases it was observed that the longitudinal Kerr ellipticity signal (which is usually used for the MO magnetometry in UHV to compensate linear birefringence in viewports) varies linearly with the total magnetic film thickness even up to at least 5 nm (for Fe/Ag) or even up to 10 nm (for Co/Cu) of the total multilayer thickness.

Let us then consider an ultrathin system grown on a nonferromagnetic substrate which is composed of two films made of the same ferromagnetic material and separated by a nonferromagnetic interlayer. When neglecting some possible interface effects[4–7] then on a basis of the above-mentioned findings one expects that the individual contributions from the magnetic films to the MO Kerr angles (i.e., the Kerr rotation or the ellipticity) in magnetic saturation should be simply proportional to their thicknesses. Consequently, when both ferromagnetic films have equal thicknesses but are magnetized antiparallel to each other, their contributions should have the same magnitudes but opposite signs. Therefore, they should cancel each other.

However, it has been recently demonstrated that when carrying out MO magnetometry experiments in UHV on antiferromagnetically exchange-coupled Fe/Cr/Fe sandwiches grown on GaAs(001) substrates, the results are in clear disagreement with the above-mentioned additivity principle.[8] More precisely, it was observed in the longitudinal Kerr ellipticity signal that the thicker bottom Fe layer (i.e., the one facing the GaAs substrate) gives a much smaller contribution to the signal than the thinner top Fe does. Consequently, when the thicker layer magnetization is oriented along the applied magnetic field, then in moderate fields where magnetizations of the both films are still antiparallel, the Kerr ellipticity signal is dominated by the contribution of the thin-

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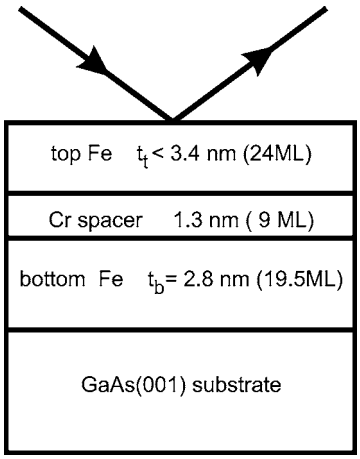

FIG. 1. A structure of the studied specimens.

ner top layer magnetized in an opposite direction with respect to the applied field. This is why the net ellipticity at positive fields has an opposite sign with respect to that measured on the Fe/GaAs(001) system. This effect results in the observation of "reversed" hysteresis loops.

In this article we discuss why the usually used additivity law is not working in such a case. In order to describe quantitatively the effect observed before,[8] we performed a more complete and precise experimental study of the MO response of the Fe/Cr/Fe systems on GaAs(001). For a fixed thickness of the bottom layer t_b, the thickness of the top one t_t was varying in a thickness range large enough to explain conditions under which the reversed loops can be observed. The experimental data were interpreted by using the electromagnetic theory for MO effects. A very good quantitative agreement between the experimental results and the theoretical calculations was achieved. In the following, the experimental details are first presented. Then the theoretical considerations are developed for explaining the experimental observations.

II. EXPERIMENT

The details of sample preparation were described elsewhere.[8] Briefly, the Fe/Cr/Fe sandwiches were grown epitaxially on clean GaAs(001) surfaces with Ga-terminated (4×6)-like reconstruction. The Fe and Cr films were deposited at room temperature by electron-beam evaporation at rates from 1 to 1.5 monolayer (ML)/min and at pressures below 2×10^{-10} mbar. Good crystallographic order was confirmed by low-energy electron diffraction and high-resolution transmission electron microscopy. The structure of the specimens is schematically shown in Fig. 1. The thickness of the bottom Fe layer t_b was kept at 2.8 nm for all specimens, while that of the top Fe layer t_t was varied from 1 to 3.4 nm. The Cr spacer thickness was set to 1.3 nm for which the Fe films are antiferromagnetically exchange coupled.[9] The maximum thickness of the sandwich structures was 7.5 nm, which still quite well satisfies conditions of the ultrathin approximation, in agreement with Refs. 2 and 3.

The MO experiments were performed *in situ* and the films were magnetized in plane along the Fe[110] azimuth which is an easy axis in this system.[10] The angle of incidence

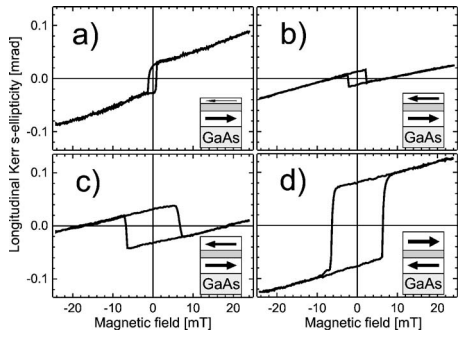

FIG. 2. Examples of magneto-optical longitudinal Kerr ellipticity hysteresis loops for *s*-polarized light with a 1.85-eV photon energy, incident at 49° on the Fe(t_t)/Cr(1.3 nm)/Fe(t_b=2.8 nm)/GaAs(001) structures. The top layer thicknesses t_t are (a) t_t=1.0 nm, (b) t_t=1.8 nm, (c) t_t=2.6 nm, and (d) t_t =3.4 nm. The insets of the individual panels indicate a structure of the specimens. The black arrows inside the Fe layers show the orientations of magnetization in remanence after applying the maximum positive field of 24 mT.

of *s*-polarized laser beam with a photon energy of 1.85 eV, corresponding to the wavelength of 670 nm, was 49°.

Typical hysteresis loops measured for different thicknesses of the top layer are shown in Fig. 2. When thickness t_t is much smaller than t_b, the hysteresis loop is regular, i.e., the Kerr ellipticity angle in positive fields has the same sign as when the top Fe layer is absent. This is demonstrated in panel (a) for t_t=1.0 nm. For thicker top layers which are anyway still thinner than the bottom Fe film ($t_t<t_b$) we start to observe that the hysteresis loop is suddenly inverted, as shown in panels (b) and (c) of Fig. 2. Note that for the loop in panel (b) the quotient of thicknesses is t_t/t_b=0.64. However, the contribution of the top Fe layer to the Kerr ellipticity already clearly dominates. When t_t is further increased so that $t_t>t_b$, the loops become regular again, as shown in panel (d) of Fig. 2 for t_t=3.4 nm. This is because the magnetization vectors for both Fe layers are switched to the inverted orientation [see the inset of panel (d)] with respect to the case of $t_t<t_b$ [insets of panels (a)–(c)]. This quite "unusual" behavior of an ultrathin system containing two ferromagnetic layers clearly demonstrates that magneto-optics is depth sensitive.

III. THEORETICAL BACKGROUND

Depth sensitivity of magneto-optics was first pointed out by Hubert and Traeger[11] for thick films. It is related to phase differences between MO signals originating in different depths of the structure. For ultrathin films these differences are, however, quite small and can be neglected in many cases. Their detection and analysis usually require an application of very careful approaches.[12]

The magneto-optical depth sensitivity to signals from individual ferromagnetic layers depends on the structure of the stratified system (i.e., on the layer thicknesses and all materials involved), on the experimental geometry (i.e., on a polarization and on an incidence angle of the incoming optical beam), and on the energy of probing photons. Here we focus on the analysis of depth sensitivity in the Fe/Cr/Fe/GaAs(001) system at our experimental geometry which uses *s*-polarized light incident at 49° from a surface

normal. The experimental data were acquired at a fixed photon energy (1.85 eV), whereas the theoretical calculations were performed as a function of photon energy. This is because these data are of interest when considering efficiency of different light sources which may be used in the magnetometry experiments.

Individual contributions of both Fe layers, indicated here by the subscript j, to the total complex MO Kerr angle (which are in general different for s-polarized and p-polarized incident waves) write

$$\Phi_j = |\Phi_j|\exp(\mathrm{i}\psi_j) = \Re(\Phi_j) + \mathrm{i}\Im(\Phi_j) \approx \theta_j - \mathrm{i}\epsilon_j. \quad (1)$$

Here Φ_j is the complex MO Kerr angle originating in the bottom layer ($j=b$) or in the top layer ($j=t$), $|\Phi_j|$ is called the Kerr magnitude, and ψ_j is the Kerr phase. Physically, ψ_j is a relative phase between the wave generated in the MO interaction (i.e., the wave polarized perpendicularly to the incident one) and the isotropically reflected incident wave. This phase can be additionally changed by a phase shifter (i.e., a phase plate or a compensator).[13] The quantities $\theta_j=\Re(\Phi_j)$ and $\epsilon_j=-\Im(\Phi_j)$ are, respectively, the Kerr rotation and the ellipticity angles.

All MO calculations were performed by using a method employing the 4×4 matrix formalism of Yeh[14] based on the electromagnetic theory of Maxwell, and applied on absorbing structures by Višňovský.[15] This calculation starts with finding four eigenmodes by solving a wave equation in each layer characterized by optical and MO (only for the selected contributing Fe layer) parameters of the corresponding materials. The eigenmodes were determined with the help of Snell's law according to which the wave vector has the same in-plane components in all layers. These components are mainly given by the angle of incidence. The amplitudes and phases of the eigenmodes at every interface must satisfy the boundary conditions for the field components. This "sewing" procedure provides relations between the incident, reflected, and transmitted waves by a linear transformation characterized by a 4×4 matrix which allows to calculate a polarization state of the reflected wave. The MO contributions of the individual layers can be quite precisely estimated by approximative approaches, taking into account also the effect of the nonferromagnetic film as will be discussed elsewhere.[16] The calculations were performed with the optical constants of Fe and Cr published by Johnson and Christy.[17] Those of GaAs were measured by Theeten *et al.*[18] and Palik.[19] The magneto-optical parameters of Fe were obtained from the spectra of the polar and longitudinal Kerr effects, on a thick polycrystalline Fe film,[20] which for the polar case also appears in Ref. 21, and the optical parameters.[17]

IV. DISCUSSION

The results for the system of Fig. 1 with antiferromagnetically coupled Fe films are shown in Fig. 3. The solid line plots the theoretical variation of the Kerr ellipticity as a function of the top Fe layer thickness t_t, as obtained from the 4×4 matrix model described above. These model data are compared with the experimental results represented by circles. Because the hysteresis loops are squares (see Fig. 2), we plot there the values of Kerr ellipticity in remanence. These values correspond to a fixed orientation of magnetization of the bottom Fe layer. This means that when t_t increases above t_b, the sign of the remanent ellipticity was inverted. From Fig. 3 it is seen that the longitudinal Kerr ellipticity for the incident s polarization is within our experimental precision, a linear function of t_t. This is in agreement with the theoretical expectation. The ellipticity signal changes sign at $t_t \approx 1.6$ nm in contrast to ≈ 2.7 nm, as would be expected from the additivity law.[2]

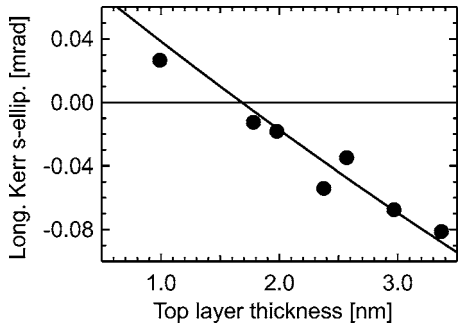

FIG. 3. Longitudinal MO Kerr ellipticities for incident s polarization in the Fe(t_t)/Cr(1.3 nm)/Fe(t_b=2.8 nm)/GaAs(001) structures as a function of the top layer thickness t_t for the positive magnetization of the bottom layer. The experimental ellipticity values (circles) are obtained from the experimental hysteresis loops measured by using s-polarized light with a 1.85-eV photon energy, incident at 49°. They are compared with a model curve calculated from the electromagnetic theory.

When comparing the experimental and theoretical results we should bear in mind that the optical and MO parameters determined on bulk specimens may differ from those for ultrathin films. Moreover, the model considers a steplike optical profile corresponding to the sharp interfaces between layers. These effects may be responsible for very small differences between the experimental data and the theory. We can therefore conclude that the electromagnetic model describes the experimental data very well.

We now turn our attention to the question of why the MO additivity law[2] works for the Kerr ellipticity angle on structures prepared on Cu or Ag substrates while it does not give acceptable results for the Fe/Cr/Fe system grown on GaAs(001). As mentioned before, the answer follows from the fact that magneto-optics is depth sensitive. This means that the MO signals Φ_j [Eq. (1)] originating in the layers located in different depths of the ultrathin structure have different phases ψ_j. For a certain phase the MO signal coming from one of the layers may cancel so that its remagnetization process will not be optically detected.

For a MO setup measuring the Kerr ellipticity we monitor $\epsilon_j=-\Im(\Phi_j)=-|\Phi_j|\sin\psi_j$. This will cancel for $\psi_j=n\pi$, where n is an integer number and $j=b$ for the bottom magnetic layer and $j=t$ for the top one. This cancellation effect could be successfully used for depth-selective observation of magnetic domains[22] or for magnetometric experiments.[12,23] If the phases ψ_j are close to the cancellation angle $n\pi$, then the layer for which the phase ψ_j is closer to $n\pi$ will contribute less to the overall MO Kerr ellipticity signal. This is actually the case of the Fe/Cr/Fe system prepared on

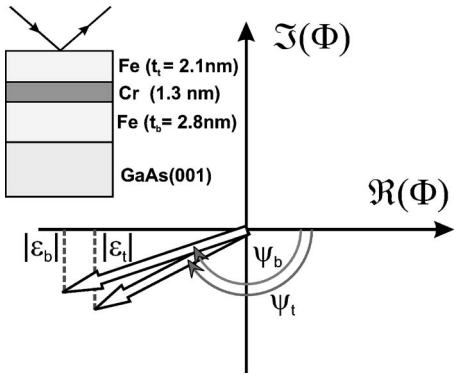

FIG. 4. Contributions of the individual top (t) and bottom (b) Fe layers to the complex longitudinal Kerr effect Φ in the system Fe(2.1 nm)/Cr(1.3 nm)/Fe(2.8 nm)/GaAs(001). The vertical dashed lines marked as $|\epsilon_t|$ and $|\epsilon_b|$ indicate, respectively, individual contributions of the top and the bottom layers to the overall Kerr ellipticity signal calculated for the positive orientation of magnetization in both Fe layers.

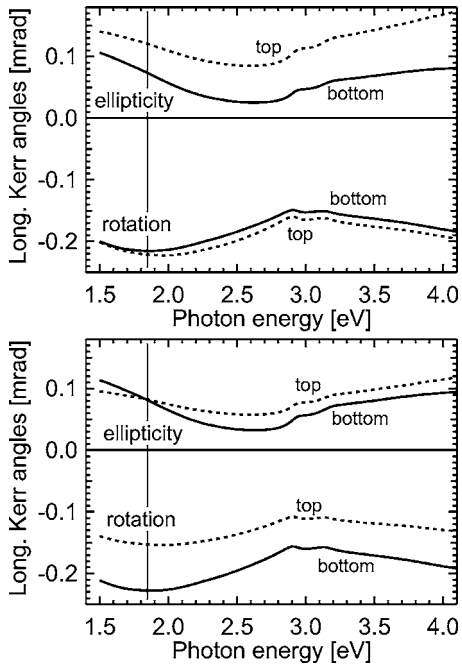

FIG. 5. Contributions of the individual Fe layers to the longitudinal magneto-optical angles, Kerr rotation, and Kerr ellipticity, as a function of photon energy in the structures (upper panel) Fe(2.6 nm)/Cr(1.3 nm)/Fe(2.8 nm)/GaAs(001) and (lower panel) Fe(1.7 nm)/Cr(1.3 nm)/Fe(2.8 nm)/GaAs(001). The contributions of the thicker bottom layers to the magneto-optical observables are plotted as solid lines while those of the top thinner layers as dashed lines. The data were calculated for *s*-polarized light incident at 49° from the surface normal. The thin vertical lines at 1.85 eV indicate the energy of photons used in the present experiments.

GaAs(001), as demonstrated in Fig. 4. This diagram shows, in agreement with a presumption, that $|\Phi_b|$ (for t_b=2.8 nm) is larger than $|\Phi_t|$ (for t_b=2.1 nm). In the first approximation their ratio is $|\Phi_b/\Phi_t| \approx t_b/t_t$. But the corresponding ellipticity signals behave differently and one gets $|\epsilon_b| < |\epsilon_t|$ because $|\Phi_b \sin \psi_b| < |\Phi_t \sin \psi_t|$. This discrepancy would disappear when measuring the Kerr rotation angles $\theta_j = \Re(\Phi_j) = |\Phi_j| \cos \psi_j$. Here we would get $|\theta_b| > |\theta_t|$ and their ratio would be roughly given by the quotient of the related thicknesses.

In other words, the Kerr magnitudes $|\Phi_j|$ characterize the total MO activity of the individual layers while the Kerr phases ψ_j determine a distribution of the $|\Phi_j|$ between the rotation and the ellipticity angles. As seen from Fig. 4, in the Fe/Cr/Fe/GaAs system the Kerr phase $\psi_j \approx -0.85\pi$. The ellipticity angle $\epsilon_j \approx -|\Phi_j| \sin \psi_j$ is therefore much more sensitive to small changes of ψ_j in contrast to the rotation angle θ_j.

It turns out that for a given material composing the ferromagnetic films the phase angles ψ_j of both layers at the given MO geometry depend mainly on the optical properties of the substrate.[16] The spacer layer affects mostly the difference $\psi_b - \psi_t$, but not the average phase. The effect of the substrate on the average phase is very important. For example, our experiments in the Fe/Cr/Fe structures would provide very similar results if the GaAs substrate was replaced by Si, which at 1.85 eV has the optical properties similar to those of GaAs.

In contrary, in the systems grown on noble-metal substrates, like Cu or Ag, it is found that for the same photon energy the typical values of the Kerr phase ψ_j are close to $-\pi/2$. In this case the roles of the rotation and the ellipticity angles are interchanged. As a result, the ellipticity angle in these systems satisfies the additivity principle quite well.[2,3] However, in this case there would be problems with the additivity law for the rotation angle.

It is also important to mention that the relative sensitivity of the MO Kerr angles to magnetizations of the individual layers depends on the energy of probing photons. This is demonstrated in Fig. 5 which shows contributions of the top and the bottom Fe layers in the Fe/Cr/Fe/GaAs structures to the MO angles as a function of photon energy. For the top panel the top Fe layer thickness is t_t=2.6 nm, while for the bottom panel t_t=1.7 nm. The bottom Fe layer thickness t_b is 2.8 nm in both cases. First, these calculated variations show that the relative contributions of both layers depend in general on the photon energy. For t_t=2.6 nm (i.e., t_t/t_b=0.93) both layers contribute to the rotation angle almost equally although the bottom layer is slightly thicker. This is partly due to the fact that the values $|\psi_j|$ differ from π and also partly due to a small departure of the system from the ultrathin approximation. The situation is, however, very different for the ellipticity angle. We indeed see that for the photon energy of 1.85 eV used in the present experiments $|\epsilon_t/\epsilon_b| = 1.64$, while at 2.7 eV this ratio increases even to $|\epsilon_t/\epsilon_b| = 3.34$. When the top layer thickness is further decreased to 1.7 nm (i.e., t_t/t_b=0.61), the spectral variation of relative sensitivities of the ellipticity to magnetizations of the bottom and of the top Fe layers is even more remarkable as it is seen on the bottom panel of Fig. 5. For the ellipticity angles at 1.85 eV we have $|\epsilon_t/\epsilon_b| = 1$, while at 2.7 eV this ratio is $|\epsilon_t/\epsilon_b| = 1.77$. This result may look quite surprising because the top film with a thickness being only about 60% of the bottom one gives an almost doubly large contribution to the ellipticity signal at this energy of 2.7 eV. For the rotation angles we get at 1.85 eV the ratio of $|\theta_t/\theta_b| = 0.67$ which at 2.7 eV increases to $|\theta_t/\theta_b| = 0.70$. These values are much closer to the ratio of thicknesses.

Another interesting aspect of the depth-dependent phase

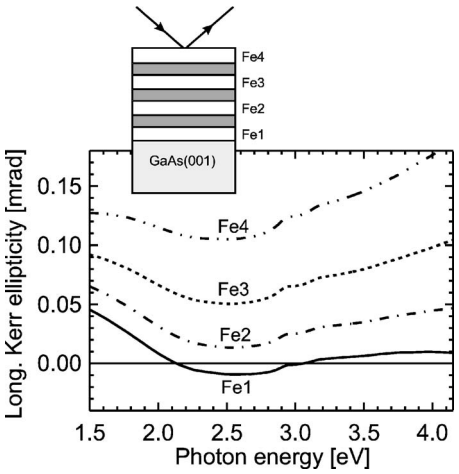

FIG. 6. Individual contributions to the longitudinal ellipticity signal in the structure Fe4/Cr/Fe3/Cr/Fe2/Cr/Fe1/GaAs(001). The thicknesses of all Fe layers are 15 ML (2.15 nm) and thicknesses of all Cr layers are equal to 9 ML (1.3 nm). The angle of incidence of the *s*-polarized photons is 49°.

effects in the structures prepared on GaAs substrates is presented in Fig. 6. It shows individual contributions to the MO Kerr ellipticity in a stack of four identical Fe layers (each with a thickness of 15 ML) separated by Cr spacers (with a thickness of 9 ML). It should be mentioned that this structure already departs from the ultrathin approximation. Similar to the previous case of the Fe/Cr/Fe/GaAs(001) systems the ellipticity signal drops rapidly with increasing depth of the film in the stack (compare with the top panel of Fig. 5). For the deepest located layer Fe1, its contribution to the ellipticity (plotted by the solid line) already crosses the zero line. When considering that the Fe layers would not be antiferromagnetically exchange coupled and each layer had a bit different coercivity, the hysteresis loop taken with the longitudinal Kerr ellipticity signal close to the photon energy of 2.5 eV would exhibit, for increasing magnetic field, three positive steps associated with the layers Fe2, Fe3, and Fe4, and one negative step related to the layer Fe1. Then, the hysteresis loop related to the layer Fe1 would be also inverted. Note that the mechanism of appearance of the inverted loop is in this case different with respect to the antiferromagnetically coupled Fe/Cr/Fe systems on GaAs. Here the ellipticity for the layer Fe1 has the opposite sign with respect to the ellipticities for layers Fe2, Fe3, and Fe4 although all four layers are magnetized in the same direction. In a Kerr magnetometry experiment this optical effect could be misinterpreted as a "negative magnetization" of the layer Fe1.

V. CONCLUSIONS

In conclusion, the present results show that in some stratified systems the Kerr effect signal does not satisfy the widely accepted additivity law.[1] As a consequence, in the Kerr MO magnetometry experiments it is, in principle, possible to misunderstand the experimental magneto-optical results when detecting only the Kerr ellipticity or only the Kerr rotation angle. It is therefore very useful to perform magnetometric experiments by using both MO signals. This allows to determine the Kerr magnitude $|\Phi|$ which should be in the best agreement with the additivity principle.[1] The Kerr magnitude can be also measured directly with a help of a phase compensator as discussed by Moog *et al.*[13] The present results on the antiferromagnetically coupled Fe/Cr/Fe systems on GaAs(001) substrates demonstrate that magnetometry data obtained on modern ultrathin structures by using the MO Kerr effect should be carefully analyzed also from the optical point of view.

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