Orientation Dependence of the Polar Kerr Effect in fcc and hcp Co

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Anisotropy of the energy dependent polar magneto-optical Kerr effect as a function of the angle between the magnetization and crystallographic axes is reported. These results are obtained on epitaxially oriented optically opaque films of hcp Co grown in two different crystallographic orientations by molecular beam epitaxy. Respective Kerr spectra of fcc Co films are found to be insensitive to the magnetization direction. This *Kerr anisotropy* is correlated with the presence of strong magnetocrystalline anisotropy in the hcp phase of Co. A perturbation theory treatment of the spin orbit coupling correctly predicts the order of magnitude of this new effect.

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The magneto-optical Kerr effect (MOKE) offers unique insight into the spin polarized electronic structure of a magnetic material because it is most strongly sensitive to those parts of the band structure which initially give rise to magnetism. Numerous recent MOKE discoveries, including quantum confinement effects [1], oscillations of the Kerr rotation with magnetic layer thickness [2], and strong correlations between MOKE and magnetic anisotropies [3], alloy chemical ordering and physical ordering [4,5], highlight its utility. In this Letter, we report a dependence of polar MOKE on the relative orientation of the magnetization and crystallographic axes in epitaxial Co films. Elemental cobalt provides an excellent model system for such studies since films greater than 1000 Å thick can be stabilized at room temperature in both the fcc and hcp phases [6] and, more importantly, in at least two crystallographic orientations for each phase. We find a strong orientation dependence of polar MOKE in epitaxial hcp Co, while analogous spectra in fcc Co are experimentally indistinguishable. This finding coincides with the presence of strong magnetocrystalline (MC) anisotropy in hcp Co, which is found to be 10 times weaker in fcc Co. A perturbation theory treatment of the spin orbit coupling predicts the correct magnitude of the orientation dependence of the Kerr effect and elucidates its relationship to the magnetic anisotropy [7].

To search for dependences of MOKE on the relative orientation of the magnetization with respect to crystallographic axes, one could choose to either measure the MOKE signal in fixed geometry [e.g., polar MOKE with magnetization **M** perpendicular ($\perp$) to the probed surface] on differently oriented surfaces of a large single crystal or change the geometry from polar (**M** $\perp$) to longitudinal (**M** $\|$) and keep the measured surface fixed. Both approaches are difficult since large single crystals are not readily available and changing the geometry from polar to longitudinal MOKE could obscure the expected subtle anisotropy effects, since additional data conversion would be required. We have therefore chosen to keep the geometry fixed and measure polar MOKE spectra on optically opaque highly oriented thin films of different orientations. To ensure that differences in sample preparation do not give rise to artifacts, our measurements have been repeated on a number of samples with different preparation methods for a given orientation [8]. The samples were 1150 Å fcc Co(110) on SrTiO$_3$(110), 1050 Å fcc Co(100) on GaAs(100), 1050 Å hcp Co(0001) on sapphire(0001) [6], 1050 Å Co(11$\bar{2}$0) on GaAs(100) [6], and 3500 Å Co(11$\bar{2}$0) on Ru(11$\bar{2}$0). All samples were checked for good epitaxial growth *in situ* using reflection high energy electron diffraction and LEED, and *ex situ* using x-ray diffraction (XRD). Structural details are summarized in Table I. All films had a single growth axis, although in-plane twinning caused the superposition of two domains (related by 180° rotation about the surface normal) in the (0001) film, and two domains (related by a 90° rotation) in the (11$\bar{2}$0) Co film on GaAs. Such in-plane twinning does not affect the *polar* MOKE effect, as discussed here. All samples showed evidence of a single crystallographic, either hcp or fcc, phase [6].

Magnetic properties were characterized by standard vibrating sample and torque magnetometry. While the room temperature saturation magnetization of 1400±50 emu/cm^3 is found to be identical within the experimental error in all cases, the MC anisotropy shows large differences between hcp and fcc Co. Results of a Fourier analysis of angle dependent torque measurements are summarized in Table II. We find a 10 times smaller MC anisotropy constant $K_1^{\mathrm{fcc}} = -(5.7 \pm 0.3) \times 10^5$ ergs/cm^3 in fcc Co, compared to $K_1^{\mathrm{hcp}} = +(4.4 \pm 0.3) \times 10^6$ ergs/cm^3 in hcp Co, consistent with previously published data [6] and with recent reports from other laboratories [9,10].

Figure 1 displays the complex polar MOKE spectra for fcc Co(110) and fcc Co(100) in the range $0.8 \leq \hbar\omega \leq 5.3$ eV [11]. There is no significant dependence of either the Kerr rotation or the Kerr ellipticity on the crystallographic orientation. The large Kerr rotation peak of 0.57° at ~1.6 eV and the relatively broad ultraviolet feature with a peak rotation of 0.45° near 3.8 eV photon

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TABLE I. Orientation, structure, and results of x-ray diffractometry of five different MBE grown Co samples.

Orientation, thicknesses, and epitaxial relationships	Peak index	Position (Å^{-1})	Corr. length (1/HWHM)	Rocking curve width (FWHM)
1150 Å Co(110)/100 Å Cu/20 Å Co/50 Å Pt/SrTiO$_3$(110)	(220)	5.0239	97 Å	1.26°
1050 Å Co(100)/100 Å Cu/20 Å Co/30 Å Pt/450 Å Ag/20 Å bcc Co/ GaAs(100)	(200)	3.5532	56 Å	1.26°
1050 Å Co(0001)/30 Å Pt/sapphire(0001)	(0002)	3.0922	200 Å	0.23°
1050 Å Co(11$\bar{2}$0)/100 Å FeCo/100 Å Fe/GaAs(100)	(11$\bar{2}$0)	5.023	>60 Å	···
3500 Å Co(11$\bar{2}$0)/Ru(11$\bar{2}$0)	(11$\bar{2}$0)	5.034	~60 Å	2.1°

energy are characteristic for these MBE fcc Co films. The ellipticity spectra show the expected Kramers-Kronig invariant behavior with a zero crossing at the infrared Kerr rotation peak. The normal incidence reflectivities, as shown in the inset, decrease monotonically with increasing energy, confirming the high sample quality [12].

Figure 2 displays the polar MOKE spectra for the two orientations of hcp Co. The Co(0001) spectrum has been reproduced numerous times and can be found in the literature [5]. To ensure the reliability of the detailed structure of the (11$\bar{2}$0) spectra, films were prepared using two completely different seeding techniques [on GaAs(100) and on Ru(11$\bar{2}$0); see Table I]. The spectra from the two (11$\bar{2}$0) films were almost identical and we display their average.

The spectra for the two Co orientations are significantly different from one another, which we attribute to the change of crystallographic orientation. Both spectra show a peak rotation of 0.43°–0.45° at ~1.3 eV photon energy and a broad ultraviolet feature with a peak rotation of 0.41° and 0.44°, centered around 3.6 and 3.9 eV, respectively. Significant differences occur throughout the measurement, and we call attention to the visible range near 2.3 eV where a minimum in the (0001) case coincides with a local maximum in the (11$\bar{2}$0) case. Even stronger effects are seen in the ellipticity spectra. The reflectivities are once again reasonably high (see inset).

We point out the strong dependence of the Kerr spectra on crystal structure. Note that the fcc and hcp Co crystal structures are both close-packed structures with equal numbers of nearest and next-nearest neighbors. The large changes we observe in the Kerr spectra in Figs. 1 and 2 highlight how sensitive MOKE is to the long-range crystal structure.

Because the *intrinsic* magnetic anisotropy arises from spin orbit coupling in the electronic structure much in the same way as MOKE, a useful comparison can be made between the presently observed *Kerr* anisotropy and *magnetocrystalline* anisotropy. Note that the fcc samples show a 10 times weaker MC anisotropy, which correlates with the absence of an orientation dependence of the respective Kerr spectra.

In order to understand these observations, we outline a perturbation theory treatment of the spin orbit coupling. This approach was first used to explain magnetic anisotropies by Brooks and later by Fletcher [13]. We assume a uniaxial anisotropy as in hcp Co, with cubic symmetry considered as a special case of the former. For the spin orbit coupling, we shall consider only the d band electrons. The single-electron Hamiltonian is written $H = H_0 + H_{\mathrm{SO}}$, where $H_0 = -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r}) + \mathcal{E}$ and

TABLE II. Results of angle dependent torque measurements of fcc and hcp Co films in different crystallographic orientations. $L2$ and $L4$ refer to the $\sin 2\theta$ and $\sin 4\theta$ Fourier components and the angle θ is measured with respect to the [100] and [110] axes for fcc and with respect to the c axis in the case of hcp Co, respectively. $\bar{K}_1$ and $\bar{K}_2$, in units of 10^6ergs/cm^3, are the averaged first and second order anisotropy constants obtained from two films in each case. The saturation magnetization of all films was 1400±50 emu/cm^3.

Co	orient.	$L2$	$L4$	$\bar{K}_1$	$\bar{K}_2$
fcc	(100)	0	$-\frac{1}{2}K_1$	-0.57	0.13
	(110)	$-(\frac{1}{4}K_1 + \frac{1}{64}K_2)$	$-(\frac{3}{8}K_1 + \frac{1}{16}K_2)$		
hcp	(11$\bar{2}$0)	$K_1 + K_2$	$\frac{1}{2}K_2$	4.4	1.3
	(0001)	$K_1 + K_2 - 2\pi M_S^2$	$\frac{1}{2}K_2$		

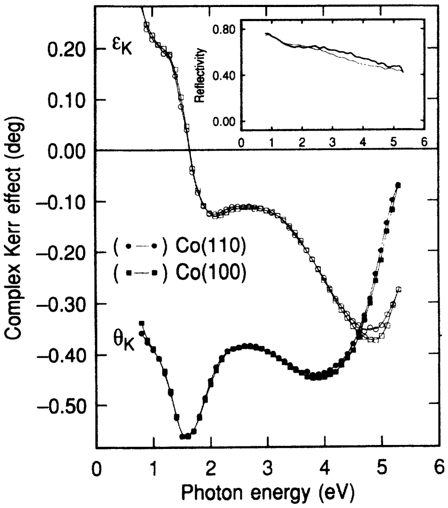

FIG. 1. Complex polar magneto-optical Kerr rotation θ_K and ellipticity ϵ_K spectra for fcc Co (110) and (100) films at room temperature and in the presence of external magnetic fields of ±20 kOe.

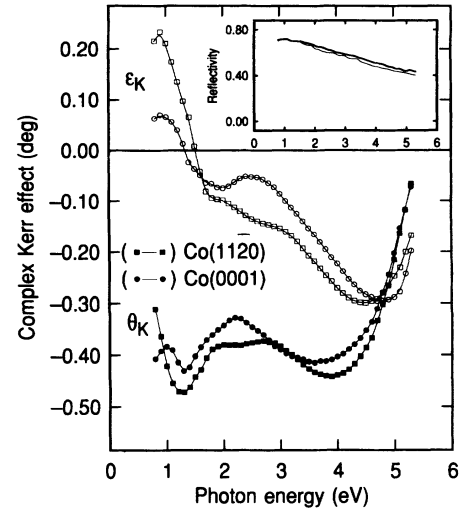

FIG. 2. Same as Fig.1, but for hcp Co films of two orientations, Co(0001) and Co(11$\bar{2}$0), and in external magnetic fields of ±28 kOe. Hcp Co shows significant variations of the Kerr spectra depending on the crystallographic orientation.

$H_{SO} = \xi\, \mathbf{L}\cdot\mathbf{S}$. Here, $\mathcal{E}$ represents the exchange splitting and ξ is the spin orbit coupling parameter. Assuming that the eigenfunctions, $\psi(0)_\alpha$, of H_0 are known, it is straightforward to deduce the form of the energy correction (to fourth order) from standard perturbation theory [14],

$$E_{SO} = \xi^2\,[E_0 + E_1\cos^2\theta] + \xi^3\,E_2 + \xi^4\,[E_3 + E_4\cos^4\theta] + O(\xi^5) \qquad (1)$$

for hcp, where θ is the angle between the **c** axis and the spin-moment (i.e., magnetization) directions. The equation for fcc is found by setting $E_1 = 0$. We now see why the MC anisotropy of fcc Co is much smaller than that of hcp Co. The constant parts of E_{SO} do not give rise to magnetic anisotropy, and the first nonvanishing anisotropic term in fcc Co, $\xi^4 E_4 = K_1^{\mathrm{fcc}}$, is $O(\xi^4)$, while it is $\xi^2 E_1 = K_1^{\mathrm{hcp}}$ in hcp Co, which is $O(\xi^2)$. This gives us an estimate of the relative size of the terms in Eq. (1), since our results show about a factor of 10 change in the first anisotropy constant in hcp versus fcc Co (see Table II). Thus, in expansions such as Eq. (1), a factor of ξ^2 appears to reduce the size of a term by a factor of 10. In a completely analogous procedure we calculate the form of the perturbed wave functions, ψ_α, of H,

$$A\psi_\alpha = \psi_\alpha^{(0)} + \xi\,\psi_\alpha^{(1)} + \xi^2\,[\psi_\alpha^{(2)} + \psi_\alpha^{(3)}\cos^2\theta] + \xi^3\,\psi_\alpha^{(4)} + \xi^4\,[\psi^{(5)} + \psi^{(6)}\cos^4\theta] + O(\xi^5) \qquad (2)$$

for hcp, where A is a normalization constant and the index α, here and throughout this paper, refers to d band electrons only, and stands for the band, spin indices, and momentum vector of the electron state. The equation for fcc is found by setting $\psi_\alpha^{(3)} = 0$.

To understand how this relates to polar MOKE, consider that the linearly polarized normally incident light wave may be regarded as being comprised equally of right-hand circularly polarized (+) and left-hand circularly polarized (−) light. The Kerr rotation, θ_K, corresponds to a relative phase shift in the reflected amplitude of the two polarizations, $\theta_K = -\frac{1}{2}(\Delta_+ - \Delta_-)$ [15], where

$$\Delta_\pm = \arctan\left(\frac{-2k_\pm}{n_\pm^2 + k_\pm^2 - 1}\right). \qquad (3)$$

Although Kerr rotations can arise from differences in indices of refraction, $n_\pm$, for simplicity we focus here on the absorptive part, $k_\pm$, only. The perturbation Hamiltonian of the optical wave field is $H_{\mathrm{Op}} = (e/mc)\,\mathbf{A}\cdot\mathbf{p}$. Thus,

$$k_\pm \propto \sum_{\alpha,\beta} |\langle\psi_\alpha|\mathbf{A}_\pm\cdot\mathbf{p}|\psi_\beta\rangle|^2 \delta(E_{\alpha\beta} - \hbar\omega), \qquad (4)$$

where the sum on β is taken over p states [16], and we have considered only the contributions of interband transitions to $k_\pm$. A straightforward expansion of ψ_α according to Eq. (2) for hcp Co leads to

$$\theta_K \sim \Delta k = \xi\Delta k^{(1)} + \xi^2\,[\Delta k^{(2)} + \Delta k^{(3)}\cos^2\theta] + \xi^3\,\Delta k^{(4)} + \xi^4\,[\Delta k^{(5)} + \Delta k^{(6)}\cos^4\theta] + O(\xi^5), \qquad (5)$$

where $\Delta k^{(i)} = k_+^{(i)} - k_-^{(i)}$, and we have used $\Delta k^{(0)} = 0$, because in the absence of spin orbit coupling there is no Kerr rotation. The expression for fcc Co is found by setting $\Delta k^{(3)} = 0$. We arrive at the familiar result that to first order, magneto-optical effects are linear in ξ [17].

We also find that to second order in ξ, there is no anisotropy of the Kerr effect for fcc Co, but that there is for hcp Co. Defining the Kerr anisotropy as $(\theta_K^{\parallel c} - \theta_K^{\perp c})/\frac{1}{2}(\theta_K^{\parallel c} + \theta_K^{\perp c})$, we estimate that this ratio should be of order 20%, assuming a factor of ξ^2 corresponds to a 1/10 decrease in the magnitude of a term. Furthermore, the first nonvanishing anisotropic term in the Kerr rotation of fcc Co is $O(\xi^4)$. Thus, the ratio of the Kerr anisotropies in hcp and fcc Co should be similar to the ratios of their anisotropy energies, e.g., 10:1.

These results are borne out by our experimental measurements. Like the Kerr rotation, the Kerr anisotropy is energy dependent, and in fact passes through zero at several energies. For an order of magnitude estimate, we consider a local maximum of the Kerr anisotropy in hcp Co at, e.g., 2.3 eV and find the anisotropy to be ~20%, in good agreement with our prediction. It is even somewhat larger below 1 eV. In fcc Co, any asymmetry present is below the experimental accuracy, consistent with the theoretical result that the Kerr anisotropy is 10 times smaller in fcc Co than in hcp Co.

Perturbation theory also predicts similar asymmetries in many other observables [14]. For example, a simple calculation of the orbital moment $\langle L_\zeta\rangle = \sum_\alpha \langle\psi_\alpha|L_\zeta|\psi_\alpha\rangle$

for hcp Co using the wave functions developed in Eq. (2) yields an equation which is perfectly analogous to Eq. (5), where the constants $\Delta k^{(i)}$ are replaced by constants $L^{(i)}$ (similarly for fcc Co). Thus, in principle, a measure of the orbital moment anisotropy is accessible from measurements of Kerr anisotropy. From the present crude analysis of the Kerr spectra for hcp Co, $\langle L_\zeta \rangle$ has an anisotropy of order 10%–20% of its average magnitude [18].

In the above discussion, we have paid close attention to the anisotropy in the Kerr effect, but little to the even larger differences between the Kerr spectra of fcc and hcp Co. In terms of the above treatment, changing from one crystal structure to the other affects the unperturbed wave functions $\psi_\alpha^{(0)}$. Thus it is easy to see why the spin orbit perturbations of these functions would be different, giving rise to the observed, distinctly different Kerr spectra for fcc and hcp Co.

In conclusion, Kerr anisotropy arises from spin orbit coupling and is generally to be expected in systems with strong MC anisotropy, i.e. in systems with low symmetry. These effects are not restricted to polar MOKE as discussed here and should also be observable in longitudinal or transverse MOKE experiments.

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