

# Electronic theory for the nonlinear magneto-optical response of transition metals at surfaces and interfaces: Dependence of the Kerr rotation on the polarization and magnetic easy axis

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We extend our previous study of the polarization dependence of the nonlinear optical response to the case of magnetic surfaces and buried magnetic interfaces. We calculate for the longitudinal and polar configuration the nonlinear magneto-optical Kerr rotation angle. In particular, we show which tensor elements of the susceptibilities are involved in the enhancement of the Kerr rotation in nonlinear optics for different configurations and we demonstrate by a detailed analysis how the direction of the magnetization and thus the easy axis at surfaces and buried interfaces can be determined from the polarization dependence of the nonlinear magneto-optical response, since the nonlinear Kerr rotation is sensitive to the electromagnetic field components instead of merely the intensities. We also prove from the microscopic treatment of spin-orbit coupling that there is an intrinsic phase difference of  $90^\circ$  between tensor elements which are even or odd under magnetization reversal in contrast to linear magneto-optics. Finally, we compare our results with several experiments on Co/Cu films and on Co/Au and Fe/Cr multilayers. We conclude that the nonlinear magneto-optical Kerr-effect determines uniquely the magnetic structure and in particular the magnetic easy axis in films and at multilayer interfaces.

## I. INTRODUCTION

Low-dimensional magnetic structures such as surfaces, thin magnetic films, and multilayer sandwiches have recently become an exciting new field of research and applications.<sup>1</sup> Especially thin magnetic films and multilayers exhibit a rich variety of properties not previously found in bulk magnetism such as enhanced or reduced moments,<sup>2</sup> oscillatory exchange coupling through nonmagnetic spacers,<sup>3-5</sup> giant magneto-resistance,<sup>6</sup> and spin-polarized quantum well states.<sup>7,8</sup> In particular, the observation of a perpendicular easy axis has attracted a great deal of interest, since this phenomenon, which cannot occur at bulk surfaces, leads to enhanced stray fields implying small magnetic domains. These small domain structures in turn may be applied for high-density magnetic recording like "perpendicular recording" devices. Thus, it is of considerable importance to characterize the easy axis of thin magnetic films and multilayer structures with buried interfaces by destructionless remote sensing. Conventional techniques for probing magnetic anisotropy are usually bulk probes such as ferromagnetic resonance<sup>9</sup> or the magneto-optic Kerr effect<sup>10,11</sup> (MOKE) deducing the magnetic surface signal from an overall bulk signal background which requires the absence of magnetism in the remainder of the sample. In a multilayer situation, however, with several magnetic layers present, these probes are inadequate to measure the magnetic signal from buried interfaces.<sup>12</sup> At present, the nonlinear magneto-optical Kerr-effect<sup>13-16</sup> (NOLIMOKE) provides a unique and destructionless tool for the probing of buried interface magnetism by remote sensing.<sup>17,18</sup>

In previous theoretical studies we calculated the nonlinear magneto-optical Kerr *spectra* by an electronic theory and showed in detail that they are fingerprints of the structural, electronic, and magnetic properties of surfaces, interfaces, and films.<sup>19</sup> Very importantly, we proposed on the basis of a detailed calculation for the longitudinal configuration with in

plane magnetization and *p* polarization of the incident light that the Kerr rotation in nonlinear optics is generally enhanced by one order of magnitude, since in nonlinear optics there is no suppression of the rotation by nonmagnetic excitations in contrast to linear optics.<sup>20</sup> This fundamental difference results from the nonlinear polarization entering the wave equation as the source term for second harmonic generation (SHG) thus making this equation inhomogeneous rather than just rendering the homogeneous wave equation at the doubled frequency. Therefore, NOLIMOKE reveals the magnetism of surfaces, interfaces, thin films, and multilayers much more drastically than linear MOKE does for bulk magnetism. Already several experiments confirmed our theory and the sensitivity of NOLIMOKE for surface and interface magnetism, and in particular the drastic enhancement of the nonlinear Kerr rotation.

Hence, in this paper, we extend our calculation of the nonlinear Kerr rotation to arbitrary Kerr configurations with arbitrary angles of incidence, arbitrary input polarization and arbitrary polarization of the reflected second harmonic (SH) light: We analyze the case of in-plane magnetization as well as magnetization perpendicular to the interface, in order to show how the strength of this enhancement of the nonlinear Kerr rotation depends on the Kerr configuration, the polarization of the incoming fundamental and outgoing SH light, and on the direction of the magnetization vector and thus on the *symmetry* of the nonlinear magneto-optical susceptibility tensor. We use then the results to propose a method using NOLIMOKE for the determination of the magnetization direction and hence the magnetic easy axis at interfaces, and the spin configuration in multilayer sandwiches.

Note, in our previous calculation<sup>21,22</sup> of the symmetry dependence of SHG the ferromagnetism of the transition metals has been neglected. Thus, in this paper, we extend our previous theory for the polarization dependence of SHG at noble and, in particular, transition-metal surfaces by including the symmetry properties of NOLIMOKE imposed by

magnetism and thus (i) to calculate the polarization dependence of the nonlinear Kerr rotation, and (ii) to determine the direction of the magnetization vector at interfaces of films and multilayers. In view of our previous results for the polarization dependence of SHG we expect an interesting dependence of the enhancement of the nonlinear Kerr rotation on the polarization and the easy axis. Note, the polarization is controlled by the matrix elements and thus it depends sensitively on the symmetry of the wave function.<sup>21,23</sup>

The paper is organized as follows: In Sec. II we present our theory for the nonlinear Kerr rotation for various configurations and in Sec. III we discuss our results with respect to the determination of the easy axis from the nonlinear Kerr effect and the magnetic phase shift in nonlinear optics. Finally, we summarize our work.

## II. THEORY

We begin our theory by the derivation of the nonlinear Kerr angle using the wave equation

$$\begin{aligned} \nabla \times \nabla \times \mathbf{E}^{(2\omega)} + \frac{\varepsilon(2\omega)}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}^{(2\omega)} \\ = - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}^{(2\omega)}, \end{aligned} \quad (1)$$

with vacuum permittivity  $\varepsilon_0$  and the nonlinear polarization

$$\frac{1}{\varepsilon_0} \mathbf{P}^{(2\omega)} = \chi^{(2)}(2\omega) : \mathbf{E}^{(1)}(\omega) \cdot \mathbf{E}^{(1)}(\omega) \quad (2)$$

as a source term. Furthermore, the law of reflection is used and the complex field amplitude  $\mathbf{E}_{r,a}^{(2\omega)}$  is decomposed into left- and right-handed circularly polarized light  $\mathbf{E}_{r,a}^{(\pm)(2\omega)}$  due to the magnetic birefringence. Then, one obtains for the complex Kerr rotation in nonlinear optics with real part  $\phi_K^{(2)}$  and ellipticity  $\varepsilon_K^{(2)}$

$$\tan \psi_K^{(2)} = \tan(\phi_K^{(2)} + i\varepsilon_K^{(2)}) = i \frac{\mathbf{E}_{r,a}^{(+)(2\omega)} - \mathbf{E}_{r,a}^{(-)(2\omega)}}{\mathbf{E}_{r,a}^{(+)(2\omega)} + \mathbf{E}_{r,a}^{(-)(2\omega)}}. \quad (3)$$

Using for the nonlinear susceptibilities a similar decomposition

$$\chi_{ijk}^{(2)\pm} = \chi_{ijk,0}^{(2)\pm} \pm \chi_{ijk,1}^{(2)} \quad (4)$$

with  $\chi_{ijk,0}^{(2)}$  and  $\chi_{ijk,1}^{(2)}$  referring to the nonlinear tensor elements being even or odd under magnetization reversal, respectively, one way rewrite Eq. (3) for not too large (but appreciable) nonlinear Kerr rotations as

$$\phi_K^{(2)} \approx \text{Re} \frac{E_\varphi^{(2\omega)}(s\text{-SH})}{E_\varphi^{(2\omega)}(p\text{-SH})}. \quad (5)$$

Here,  $E_\varphi^{(2\omega)}(s\text{-SH})$  and  $E_\varphi^{(2\omega)}(p\text{-SH})$  denote the reflected SH field amplitudes polarized perpendicular ( $s$ ) to or in the optical plane ( $p$ ), respectively, and both resulting from incident light of polarization angle  $\varphi$ . Using electro-dynamical theory these fields  $E_\varphi^{(2\omega)}$  are expressed by the nonlinear susceptibilities, which are then determined by electronic theory. Note that in the subsequent analysis we will take into ac-

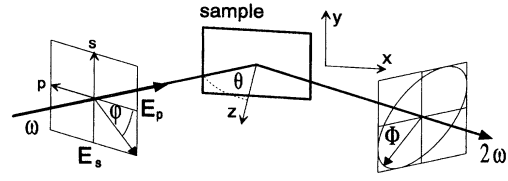


FIG. 1. The polarization and geometry of the incoming  $\omega$  and reflected  $2\omega$  light, respectively.  $\varphi$  and  $\Phi$  are the polarizations of the incident light and the reflected frequency-doubled photons.  $\varphi=0^\circ$  corresponds to  $p$  polarization and  $\varphi=90^\circ$  to  $s$  polarization.  $\theta$  denotes the angle of incidence. The crystal axes  $x$  and  $y$  are in the crystal-surface plane whereas  $z$  is parallel to the surface normal.

count *all* nonvanishing nonlinear tensor elements of the longitudinal and polar Kerr configurations.

The above formula Eq. (5) for the Kerr rotation in nonlinear optics follows (for not too large nonlinear Kerr rotations) also from the expression given by Koopmans and Rasing.<sup>24</sup> To calculate the fields  $E_\varphi^{(2\omega)}(s\text{-SH})$  and  $E_\varphi^{(2\omega)}(p\text{-SH})$ , we extend our previous work on the symmetry properties and polarization dependence of optical SHG to the magnetic case. The reflected light at frequency  $2\omega$  is given as shown by Böhmer *et al.*<sup>21,22</sup> for (001) surfaces and interfaces of cubic (fcc or bcc) crystals (with  $z$  being the surface normal and  $C_{4v}$  symmetry group) without magnetization<sup>25</sup> by

$$\begin{aligned} E^{(2\omega)}(\Phi, \varphi) = 2i \left( \frac{\omega}{c} \right) |E_0^{(\omega)}|^2 \begin{pmatrix} A_p F_c \cos \Phi \\ A_s \sin \Phi \\ A_p N^2 F_s \cos \Phi \end{pmatrix} \\ \times \begin{pmatrix} 0 & 0 & 0 & | & 0 & \chi_{xzx}^{(2)} & 0 \\ 0 & 0 & 0 & | & \chi_{xzx}^{(2)} & 0 & 0 \\ \chi_{xzx}^{(2)} & \chi_{xzx}^{(2)} & \chi_{xzz}^{(2)} & | & 0 & 0 & 0 \end{pmatrix} \\ \times \begin{pmatrix} f_c^2 t_p^2 \cos^2 \varphi \\ t_s^2 \sin^2 \varphi \\ f_s^2 t_p^2 \cos^2 \varphi \\ 2f_s t_p t_s \cos \varphi \sin \varphi \\ 2f_c f_s t_p^2 \cos^2 \varphi \\ 2f_c t_p t_s \cos \varphi \sin \varphi \end{pmatrix}. \end{aligned} \quad (6)$$

Here,  $\Phi$  and  $\varphi$  denote the angles of polarization of the reflected frequency doubled and of the incident light (see Fig. 1).  $f_{c,s}$  and  $F_{c,s}$  are the Fresnel coefficients and  $t_{s,p}$  and  $T_{s,p}$  are the linear transmission coefficients for the fundamental and frequency doubled light. The complex indices of refraction at frequencies  $\omega$  and  $2\omega$  are  $n=n_1+ik_1$  and  $N=n_2+ik_2$ . The Fresnel factors are

$$f_s = \sin \theta / n, \quad f_c = \sqrt{1 - f_s^2},$$

and

$$F_s = \sin \Theta / N, \quad F_c = \sqrt{1 - F_s^2}, \quad (7)$$

where  $\theta$  and  $\Theta$  denote the angle of incidence of the incoming light and the angle of reflection of the reflected SHG light, respectively. The linear transmission coefficients are given by<sup>21,22,26</sup>

$$t_p = \frac{2 \cos \theta}{n \cos \theta + f_c}, \quad t_s = \frac{2 \cos \theta}{\cos \theta + n f_c},$$

$$T_p = \frac{2 \cos \Theta}{N \cos \Theta + F_c}, \quad T_s = \frac{2 \cos \Theta}{\cos \Theta + N F_c}. \quad (8)$$

The corresponding amplitudes  $A_p$  and  $A_s$  in Eq. (6) are

$$A_p = \frac{2 \pi T_p}{\cos \Theta}, \quad A_s = \frac{2 \pi T_s}{\cos \Theta}, \quad (9)$$

respectively. It is interesting to note that the prefactor  $\delta z$  introduced by Böhmer<sup>22</sup> is absorbed in the tensor  $\chi_{ijk}^{(2)}$ . The relationship of this factor with the skin depth is discussed in Appendix A.

Combining now Eqs. (5) and (6) we determine the Kerr rotation and its dependence on  $\chi_{ijk}^{(2)}$  for various configurations. First we consider the *longitudinal configuration* with magnetization parallel to the interface,  $\mathbf{M} \parallel \hat{x}$ , and where the optical plane is the  $xz$  plane. In this configuration, the nonlinear susceptibility tensor contains ten different nonvanishing tensor elements with five of them being even under magnetization reversal and the other five ones being odd.<sup>14</sup> Thus, the symmetry breaking by the magnetization induces five more tensor elements and causes the nonmagnetic ones to become all different. The nonlinear susceptibility tensor is given explicitly by

$$\begin{pmatrix} 0 & 0 & 0 & | & 0 & \chi_{xxz}^{(2)} & \chi_{xxy}^{(2)} \\ \chi_{yxx}^{(2)} & \chi_{yyy}^{(2)} & \chi_{yzz}^{(2)} & | & \chi_{yyz}^{(2)} & 0 & 0 \\ \chi_{zxx}^{(2)} & \chi_{zyy}^{(2)} & \chi_{zzz}^{(2)} & | & \chi_{zyz}^{(2)} & 0 & 0 \end{pmatrix}. \quad (10)$$

In Appendix B we give the general expressions resulting from this tensor for the reflected  $p$ - and  $s$ -polarized SH fields generated from fields with polarization  $\varphi$ . For the calculation of the nonlinear Kerr rotation it is convenient to assume  $p$  or  $s$  polarization of the incident light and to detect the rotated polarization plane in the reflected SH signal upon magnetization reversal. Hence, one needs to know the fields for the polarization combinations referring to the incoming and outgoing light:  $p \rightarrow p$ ,  $p \rightarrow s$ ,  $s \rightarrow p$ , and  $s \rightarrow s$ . One obtains then for the longitudinal configuration for  $p$ -polarized SH light generated from  $p$ -polarized incident light

$$E_p^{(2\omega)}(p - \text{SH}) = 2i |E_0^{(\omega)}|^2 A_p t_p^2 [F_c \chi_{xxz}^{(2)} 2f_c f_s + N^2 F_s (\chi_{zxx}^{(2)} f_c^2 + \chi_{zzz}^{(2)} f_s^2)], \quad (11)$$

for  $p$ -polarized SH light generated from  $s$ -polarized incident light

$$E_s^{(2\omega)}(p - \text{SH}) = 2i |E_0^{(\omega)}|^2 A_p t_s^2 N^2 F_s \chi_{zyy}^{(2)}, \quad (12)$$

for  $s$ -polarized SH light generated from  $p$ -polarized incident light

$$E_p^{(2\omega)}(s - \text{SH}) = 2i |E_0^{(\omega)}|^2 A_s t_p^2 (\chi_{yxx}^{(2)} f_c^2 + \chi_{yzz}^{(2)} f_s^2), \quad (13)$$

and for  $s$ -polarized SH light generated from  $s$ -polarized incident light

$$E_s^{(2\omega)}(s - \text{SH}) = 2i |E_0^{(\omega)}|^2 A_s t_s^2 \chi_{yyy}^{(2)}. \quad (14)$$

Note that *magnetism* occurs in all ten nonvanishing elements of the tensor  $\chi_{ijk}^{(2)}$  and in the complex indices of refraction at the fundamental and the SH frequency,  $n(\omega)$  and  $N(2\omega)$ , respectively. The dominant nonlinear magneto-optical Kerr effect, however, results from  $\chi_{ijk}^{(2)}$ , in particular from the five tensor elements  $\chi_{xxy}^{(2)}$ ,  $\chi_{yxx}^{(2)}$ ,  $\chi_{yyy}^{(2)}$ ,  $\chi_{yzz}^{(2)}$ , and  $\chi_{zyz}^{(2)}$ , which are odd upon magnetization reversal. Using these expressions for  $E_\varphi^{(2\omega)}$  we find in the case of the longitudinal (= meridional) Kerr configuration for the nonlinear Kerr rotation of  $p$ -polarized incoming light

$$\begin{aligned} \phi_{K,p}^{(2)} &= \text{Re} \frac{E_p^{(2\omega)}(s - \text{SH})}{E_p^{(2\omega)}(p - \text{SH})} \\ &= \text{Re} \frac{A_s}{A_p} \frac{\chi_{yxx}^{(2)} f_c^2 + \chi_{yzz}^{(2)} f_s^2}{F_c \chi_{xzx}^{(2)} 2f_c f_s + N^2 F_s (\chi_{zxx}^{(2)} f_c^2 + \chi_{zzz}^{(2)} f_s^2)}, \end{aligned} \quad (15)$$

and in for  $s$ -polarized incoming light

$$\phi_{K,s}^{(2)} = \text{Re} \frac{E_s^{(2\omega)}(s - \text{SH})}{E_s^{(2\omega)}(p - \text{SH})} = \text{Re} \frac{A_s}{A_p} \frac{\chi_{yyy}^{(2)}}{N^2 F_s \chi_{zyy}^{(2)}}. \quad (16)$$

It is remarkable that the NOLIMOKE rotation measures the *electric-field vectors* rather than intensities. Note, in the case of the transverse (= equatorial) Kerr configuration ( $\mathbf{M} \parallel \hat{y}$ , optical plane is the  $xz$  plane) no Kerr rotation can be observed. Instead one measures an intensity change upon magnetization reversal,<sup>15,17</sup> whereas the total reflected SH intensity does not change upon magnetization reversal in the longitudinal configuration.<sup>24</sup>

Secondly, we determine the Kerr rotation for the *polar configuration*, in which the magnetization is perpendicular to the surface. Now, the optical plane is again the  $xz$  plane. However, in contrast to the usual notion in linear optics, perpendicular incidence is not yet assumed, since SHG behaves differently for the nonlinear excitation in the interface plane and perpendicular to it. Linear optics, however, makes no such difference.<sup>27</sup> In the case of the polar configuration the nonlinear susceptibility has seven nonvanishing tensor elements, five of which are different. Three of these elements are even and two ( $\chi_{xyz}^{(2)}$  and  $\chi_{yzx}^{(2)}$ ) are odd in  $\mathbf{M}$ . Note that the polar configuration is much more symmetric than the longitudinal Kerr configuration thus causing more tensor elements to vanish and to be equal. In detail, the nonlinear susceptibility is given by

$$\begin{pmatrix} 0 & 0 & 0 & | & \chi_{xyz}^{(2)} & \chi_{zxx}^{(2)} & 0 \\ 0 & 0 & 0 & | & \chi_{zxx}^{(2)} & -\chi_{xyz}^{(2)} & 0 \\ \chi_{zxx}^{(2)} & \chi_{zxx}^{(2)} & \chi_{zzz}^{(2)} & | & 0 & 0 & 0 \end{pmatrix}. \quad (17)$$

The general expressions resulting from this tensor for the reflected  $p$ -polarized SH field generated from  $\varphi$  input polarization and for the  $s$ -polarized SH field are again given in Appendix B. The calculation of the fields for the polarization combinations  $p \rightarrow p$ ,  $p \rightarrow s$ ,  $s \rightarrow p$ , and  $s \rightarrow s$  gives for

$p$ -polarized SH light generated from  $p$ -polarized incident light in the polar configuration

$$E_p^{(2\omega)}(p-SH) = 2i|E_0^{(\omega)}|^2 A_p t_p^2 [F_c \chi_{xxz}^{(2)} 2f_c f_s + N^2 F_s (\chi_{zxx}^{(2)} f_c^2 + \chi_{zzz}^{(2)} f_s^2)] , \quad (18)$$

for  $p$ -polarized SH light generated from  $s$ -polarized incident light

$$E_s^{(2\omega)}(p-SH) = 2i|E_0^{(\omega)}|^2 A_p N^2 F_s t_s^2 \chi_{zxx}^{(2)} , \quad (19)$$

for  $s$ -polarized SH light generated from  $p$ -polarized incident light

$$E_p^{(2\omega)}(s-SH) = -2i|E_0^{(\omega)}|^2 A_s \chi_{xyz}^{(2)} 2f_c f_s t_p^2 , \quad (20)$$

and finally for  $s$ -polarized SH light generated from  $s$ -polarized incident light

$$E_s^{(2\omega)}(s-SH) = 0. \quad (21)$$

Note that it makes no sense to consider the field  $E_{\varphi=\pi/4}^{(2\omega)}(s-SH)$  for both the polar and longitudinal Kerr configuration, since due to the magnetization one or even both of the two quantities  $E_p^{(2\omega)}(s-SH)$  and  $E_s^{(2\omega)}(s-SH)$  are non-zero in this case in contrast to the nonmagnetic case where both  $E_p^{(2\omega)}(s-SH)$  and  $E_s^{(2\omega)}(s-SH)$  vanish. Using the fields  $E_{\varphi}^{(2\omega)}$  in the *polar* Kerr configuration, we obtain for the nonlinear magneto-optical rotation in the case of  $p$ -polarized incident light

$$\begin{aligned} \phi_{K,p}^{(2)} &= \text{Re} \frac{E_p^{(2\omega)}(s-SH)}{E_p^{(2\omega)}(p-SH)} \\ &= -\text{Re} \frac{A_s}{A_p} \frac{\chi_{xyz}^{(2)} 2f_c f_s}{F_c \chi_{xxz}^{(2)} 2f_c f_s + N^2 F_s (\chi_{zxx}^{(2)} f_c^2 + \chi_{zzz}^{(2)} f_s^2)} \end{aligned} \quad (22)$$

and in the case of  $s$ -polarized incident light

$$\phi_{K,s}^{(2)} = \text{Re} \frac{E_s^{(2\omega)}(s-SH)}{E_s^{(2\omega)}(p-SH)} = \text{Re} \frac{A_s}{A_p} \frac{0}{N^2 F_s t_s^2 \chi_{zxx}^{(2)}} . \quad (23)$$

Note, in both the longitudinal and the polar Kerr configuration and for both  $p$  and  $s$  input polarization the Kerr rotation contains only odd tensor elements in the numerator and only even tensor elements in the denominator as generally expected and as was derived already by Pustogowa *et al.*<sup>20</sup> for not too large rotation angles. The dependence on the incident angle results here and in Ref. 20 exclusively from the linear optical coefficients. Assuming  $\chi_{yxx}^{(2)} < \chi_{yzz}^{(2)}$  and  $\chi_{zxx}^{(2)} < \chi_{zzx}^{(2)} < \chi_{zzz}^{(2)}$ , Eq. (15) for the nonlinear Kerr rotation in the longitudinal geometry yields in agreement with Ref. 20

$$\phi_{K,p}^{(2)} = \text{Re} \frac{A_s}{A_p N^2 F_s} \frac{\chi_{yzz}^{(2)}}{\chi_{zzz}^{(2)}} \approx \frac{1}{N \sin \theta} \frac{\chi_{yzz}^{(2)}}{\chi_{zzz}^{(2)}} , \quad (24)$$

if the same approximations are made. This fact is easily seen from the  $\sin^2 \Theta$  terms in the *even linear* tensor elements contributing to the denominator of  $\phi_{K,p-in}^{(2)}$  and cancelling the  $\sin \Theta$  originating from the *odd linear* susceptibility tensor elements. In Ref. 20, however, also the magnetism in the

linear optical factors and the detailed form of the nonlinear Fresnel coefficients belonging to the particular choice of the configuration has been included. Thus, the effects of the  $1/N \sin \theta$  term are suppressed what results in a much weaker angular dependence of  $\phi_{K,p}^{(2)}$ .

This completes then the determination of the polarization dependence of the Kerr rotation in nonlinear optics. The Kerr angle is expressed in terms of  $\chi_{ijk}^{(2)}$  which may be calculated by an electronic theory. According to Eqs. (15), (16) and (22), (23) one may determine the easy magnetic axis from the Kerr rotation. Of course, it is straightforward to also give expressions for the ellipticity  $\varepsilon_K^{(2\omega)} = \text{Im}[E_{\varphi}^{(2\omega)}(s-SH)/E_{\varphi}^{(2\omega)}(p-SH)]$ . In the next section we discuss in more detail and quantitatively the Kerr rotation in nonlinear optics at surfaces, interfaces, and in thin films. The understanding of these cases is a prerequisite for multilayers, where additional interference structures come into play.<sup>24</sup>

### III. RESULTS AND DISCUSSION

We now discuss the special cases of perpendicular and grazing incidence. For *perpendicular* incidence ( $\theta = \Theta = 0^\circ$ ) the Fresnel factors become

$$f_s = 0, \quad f_c = 1, \quad F_s = 0, \quad F_c = 1 \quad (25)$$

and the linear transmission coefficients simplify to

$$t_p = t_s = \frac{2}{1+n}, \quad T_p = T_s = \frac{2}{1+N}. \quad (26)$$

Thus, the corresponding amplitudes  $A_p$  and  $A_s$  in Eq. (6) are

$$A_p = A_s = \frac{4\pi}{1+N}. \quad (27)$$

Thus, the nonlinear Kerr rotation in the *longitudinal* configuration for  $p$ -input polarization becomes

$$\phi_{K,p-in,long.}^{(2)} = 1 \text{Re} \frac{\chi_{yxx}^{(2)} 1}{0 \chi_{zxx}^{(2)} 0 + 0} \rightarrow \infty \quad (28)$$

and for  $s$ -input polarization

$$\phi_{K,s-in,long.}^{(2)} = 1 \text{Re} \frac{\chi_{yyy}^{(2)} 1}{0 \chi_{zyy}^{(2)}} \rightarrow \infty. \quad (29)$$

Thus, the nonlinear Kerr rotation angle becomes arbitrarily large for perpendicular incidence. This is equally true for  $p$  and for  $s$  input polarization. Note, this divergence of the angle means according to Eq. (5) a rotation by up to  $90^\circ$ . We use now Eqs. (15) and (16) to calculate the nonlinear Kerr rotation angle for Fe in the longitudinal configuration for  $p$  and  $s$ -polarized incident light. Results for  $\Phi_K^{(2)}$  are shown in Fig. 2. These results were obtained from our microscopic theory<sup>20</sup> for the nonlinear Kerr susceptibilities  $\chi_{yzz}^{(2)}$  and  $\chi_{zzz}^{(2)}$ , the spin-orbit coupling constant has been kept fixed at 50 meV, and the complex indices of refraction at 1.6 and 3.2 eV were taken from Johnson and Christy<sup>28</sup> ( $n = 2.87 + i3.28$ ,  $N = 2.12 + i2.50$ ). For the absolute ratios and the relative phases of the complex tensor elements we use the values  $\chi_{zxx}^{(2)} = 0.60 \chi_{zzz}^{(2)} e^{i1.945\pi}$ ,  $\chi_{yyy}^{(2)} = \chi_{yxx}^{(2)}$

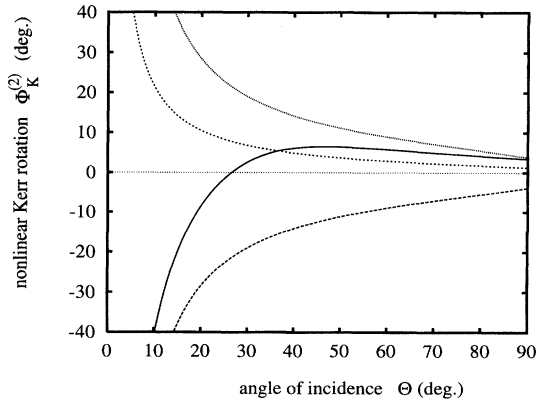


FIG. 2. Nonlinear Kerr rotation angles for  $p$ -polarized incident light  $\phi_{K,p}^{(2)}$  (full and short-dashed curves) and for  $s$ -polarized incident light  $\phi_{K,s}^{(2)}$  (long-dashed and dotted curves) for Fe at 770 nm as a function of the angle of incidence  $\theta$  in the longitudinal Kerr configuration. The relative phases between  $\chi_{zzx}^{(2)} = \chi_{zyy}^{(2)}$  and  $\chi_{zzz}^{(2)}$  is  $0.505\pi$  in the full and long-dashed curves and  $1.505\pi$  in the short-dashed and dotted curves.

$= 1.60 \chi_{zzz}^{(2)} e^{i0.5\pi(\lambda_{s.o.}/\hbar\omega)}$ , and  $\chi_{yzz}^{(2)} = \chi_{zzz}^{(2)} e^{i0.5\pi(\lambda_{s.o.}/\hbar\omega)}$ . For the remaining quantities we use  $\chi_{zxx}^{(2)} = \chi_{zyy}^{(2)} = 0.0681 \chi_{zzz}^{(2)} e^{i0.505\pi}$  for the full (  $\Phi_{K,p}^{(2)}$  ) and long-dashed (  $\Phi_{K,s}^{(2)}$  ) curves and  $\chi_{zxx}^{(2)} = \chi_{zyy}^{(2)} = 0.0681 \chi_{zzz}^{(2)} e^{i1.505\pi}$  for the short-dashed (  $\Phi_{K,p}^{(2)}$  ) and dotted (  $\Phi_{K,s}^{(2)}$  ) curves.

The results of Fig. 2 clearly show the divergence of the nonlinear Kerr rotation  $\Phi_K^{(2)}$  for  $p$ - and  $s$ -polarized incident light in the case of perpendicular incidence and the increased enhancement for  $s$  polarization. This result is supported by experiments<sup>24</sup> on Fe/Cr multilayers which find a drastic enhancement of the nonlinear Kerr rotation in the longitudinal configuration. Upon decreasing the angle of incidence from  $75^\circ$  to  $15^\circ$ , values of  $\phi_{K,p-in}^{(2)}$  from  $1^\circ$  to  $5^\circ$  and of  $\phi_{K,s-in}^{(2)}$  from  $5^\circ$  to  $15^\circ$  are observed. Thus, in contrast to linear optics, there is no need to resort to uranium based compounds, large external magnetic fields or low temperatures in order to obtain arbitrarily large Kerr rotations in nonlinear optics.

The fact that the experimental values do not increase monotonously in this range of angles of incidence is readily understood from interference effects in multilayers and is discussed in detail by Koopmans and Rasing. As pointed out by these authors, at surfaces and in films  $\phi_{K,p-in}^{(2)}$  and  $\phi_{K,s-in}^{(2)}$  should be smooth functions of angle of incidence in agreement with our result. Our result yields in addition that the absolute values and relative phases even of relatively small tensor elements are of major importance for an adequate description of the nonlinear Kerr rotation angle  $\Phi_K^{(2)}$  for  $p$ - and  $s$ -polarized incident light. Sign changes (see full curve in Fig. 2) occur even at surfaces and in thin homogeneous Fe films. Interferences in experiment do not only result from the multilayer structure, which brings additional symmetry requirements and new electronic states into play, but also from the superposition of several complex tensor elements contributing to  $\Phi_K^{(2)}$ . This might be the reason for the ‘‘calibration problem’’ quoted by Wierenga *et al.*<sup>17</sup> More work has to be done on this point both theoretically and experimentally.

Note that our theory yields the divergence of the nonlinear Kerr rotation for perpendicular incidence in the case of  $s$ - and  $p$ -polarized incident light. For  $p$  polarization, the limit of perpendicular incidence is interesting, since in this situation the excitation is parallel to the magnetization  $\mathbf{M}$ . This is due to the odd tensor element  $\chi_{yxx}^{(2)}$ , which does not vanish. It is clear that the approximation of not too large nonlinear Kerr rotations underlying our theory will break down in the case of strictly perpendicular incidence. The conclusion of this result, however, will not change. It should also be pointed out that this large enhancement of the nonlinear Kerr rotation is due to the arrangement of odd and even tensor elements in the rotation where the even ones in the denominator of the formula for the nonlinear Kerr rotation cannot be excited for perpendicular incidence due to vanishing Fresnel factors  $f_s$  for the fundamental polarization at frequency  $\omega$ .

Our theory yields that, in general, for finite angles of incidence the enhancement of the nonlinear Kerr rotation should be much more pronounced for  $s$ -polarized incident light, since in  $p$  polarization the denominator of the formula for  $\phi_{K,p}^{(2)}$  contains several independent contributions which tend to decrease  $\phi_{K,p}^{(2)}$ . Furthermore, the excitation in  $z$  direction (in  $p$  polarization) entering this denominator causes an only moderate enhancement of  $\phi_{K,p}^{(2)}$ . This is in agreement with the theoretical results by Pustogowa *et al.*<sup>20</sup> and with all experiments presently available.<sup>22,24,29</sup> Note that the microscopic theory by Pustogowa *et al.* treats in particular the enhancement in the longitudinal Kerr configuration yielding a  $\phi_{K,p-in}^{(2)}$  of  $2^\circ$  to  $4^\circ$  for a Fe surface in the optical frequency range. This prediction is experimentally confirmed by Koopmans and Rasing. It is important to make two remarks concerning the choice of the longitudinal configuration and  $p$  input polarization by Pustogowa *et al.*: (i) Only in the longitudinal configuration linear and nonlinear Kerr rotations can be meaningfully compared since for the polar configuration the nonlinearity parallel to the surface depends very much on the localization of the excited electrons and their degree of jelliumlike behavior and in the transverse configuration no rotation is observed. Only an intensity change upon magnetization reversal will happen. (ii) Only in the longitudinal configuration with incident  $p$  polarization the Kerr rotation angle can be defined as in linear optics with respect to the polarization of the incident photons at frequency  $\omega$ . For incident  $s$  polarization as well as for the polar Kerr configuration one has to resort to the definition of  $\phi_{K,p}^{(2)}$  as being one half of the angle by which the major half axis of the second harmonic polarization ellipsis is rotated upon magnetization reversal thus not referring at all to the incident beam polarization.

Note, Pustogowa *et al.* calculated the nonlinear Kerr rotation including the magnetism in the dominant tensor elements in the nonlinear as well as in the linear susceptibilities. In particular, the  $1/\sin\Theta$  dependence of  $\phi_{K,p}^{(2)}$  in the longitudinal configuration is also present in their theory in the linear Fresnel coefficients but strongly suppressed by other terms in these factors. Thus, the dependence of  $\phi_{K,p}^{(2)}$  on the angle of incidence  $\Theta$  comes out much weaker in this treatment, which is particularly suited for the frequency dependence of  $\phi_{K,p-in}^{(2)}$ . In this paper, we emphasize mostly the symmetry aspects and thus the magnetism is neglected in the linear

susceptibilities contributing to the Fresnel and transmission coefficients. This approximation is justified by the result by Pustogowa *et al.* who found the enhancement dominantly caused by the nonlinear susceptibilities. Thus, for the symmetry considerations, all the nonlinear tensor elements are included in our present paper.

In the *polar* Kerr configuration we obtain for perpendicular incidence ill-defined formulas for both *p*- and *s*-input polarization

$$\phi_{K,p-in,pol.}^{(2)} = \phi_{K,s-in,pol.}^{(2)} = \text{Re} \frac{0}{0}. \quad (30)$$

Expansion of the formulas for nearly perpendicular incidence, however, shows that there is no *s*-polarized second harmonic signal expected within the electric dipole approximation, but there is *p*-SH generated from *s* input polarization by exciting the tensor element  $\chi_{yxx}^{(2)}$ , thus leading to a vanishing  $\phi_{K,s}^{(2)}$ . On the other hand, for *p*-input polarization both *s*- (numerator of  $\phi_{K,p}^{(2)}$ ) and *p*-SH (denominator) yield become finite (apart from accidental zeros due to interference of the several complex tensor elements or due to a special choice of the frequency  $\omega$ ) thus yielding in general a finite  $\phi_{K,p}^{(2)}$ .

This is why NOLIMOKE is a unique tool for the determination of the easy axis in films and at interfaces and which cannot be determined by other tools. According to our analysis it is necessary for that purpose to shine light in at slightly off-perpendicular incidence. Then *perpendicular* interface magnetization (see our theory for *polar* Kerr configuration) would show no nonlinear Kerr rotation for *s* input but an appreciable Kerr angle for *p* input. On the other hand, the characteristic signature of *in-plane* magnetization (see our theory for *longitudinal* Kerr configuration) is expected to exhibit a moderately enhanced nonlinear Kerr angle for *p* input but a large nonlinear Kerr rotation for *s* input polarization. Thus, in switching from *s* to *p* input, the nonlinear Kerr rotation should increase for perpendicular and decrease for in plane easy axis.

Since NOLIMOKE can also distinguish the magnetic signals coming from different interfaces as proposed by Hübner *et al.*<sup>19</sup> and impressively experimentally detected by Wierenga *et al.*<sup>17,30</sup> on Co/Au sandwiches, nonlinear magneto-optics is in addition a unique and sensitive probe for the detection and investigation of spin configurations, in particular, canted spin structures generated by oscillatory exchange coupling in magnetic sandwich heterostructures. For example, in the case of canted spins in neighboring layers one may choose the light configuration such that it does not couple to the magnetization parallel to the interface, but only to the magnetization component perpendicular to the interface. Thus, then a canted spin configuration may be detected. In the case of antiparallel magnetization in neighboring thin layers 1 and 2 one has approximately for the SH yield:

$$I(\text{SH}) = I_1(M) + I_2(-M) + \dots \\ = I_1(M) + I_2(M) + [I_2(-M) - I_2(M)]. \quad (31)$$

Finally we discuss the NOLIMOKE rotation for *grazing* incidence  $\theta = \Theta = 90^\circ$ . In this case we have  $\cos\theta = 0$  and  $\sin\theta = 1$  and get the following Fresnel factors:

$$f_s = \frac{1}{n}, \quad f_c = \sqrt{1 - \left(\frac{1}{n}\right)^2}, \quad F_s = \frac{1}{N}, \quad F_c = \sqrt{1 - \left(\frac{1}{N}\right)^2}. \quad (32)$$

For grazing incidence it is meaningless to consider the transmission coefficients alone which vanish. Instead what matters for the Kerr rotation are the amplitudes which  $A_p$  and  $A_s$  which become

$$A_p = \frac{4\pi}{F_c}, \quad A_s = \frac{4\pi}{NF_c}. \quad (33)$$

This yields the ratio

$$\frac{A_s}{A_p} = \frac{1}{N}. \quad (34)$$

These equations together with Eqs. (15), (16) and (22), (23) show that the use of grazing incidence does not lead to simplified formulas for the NOLIMOKE rotation in the case of *p*- or *s*-input polarization.

Furthermore, it is an interesting observation that in NOLIMOKE there is a relative phase of  $90^\circ$  between the odd and even elements of the nonlinear susceptibility tensor  $\chi_{ijk}^{(2)}$  in contrast to linear optics. This phase has already been found in the early theories by Hübner and Bennemann<sup>13</sup> and Pan *et al.*<sup>14</sup> and has later been observed in the experiment by Wierenga *et al.*<sup>17</sup> They observed  $\Phi = 88^\circ$ . We discuss the microscopic origin of this relative phase in Appendix C. In summary, we have shown that in SH the Kerr rotation depends sensitively on the light polarization, of the magnitude and direction of the magnetization and therefore on the easy axis.

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## APPENDIX A: NONLINEAR RESPONSE DEPTH AND SKIN DEPTH

Since the nonlinear optical response results only from the range of broken electronic inversion symmetry, Böhmer *et al.* introduced in Eq. (6) an additional artificial prefactor of  $\delta z$  denoting the nonlinear response depth. In this appendix we briefly discuss the relationship of the prefactor  $\delta z$  introduced in Ref. 22 with the skin depth and show that this factor is already automatically contained in our microscopic expression of the nonlinear magneto-optical susceptibility.<sup>19</sup> Thus, in the present theory this prefactor is implicitly included in the expression given above for  $E^{(2\omega)}(\Phi, \varphi)$ , since it can be combined with the ratio  $\omega/c$  to yield the dimensionless constant  $qa$

$$\frac{\omega}{c} \times \delta z = qa, \quad (A1)$$

where  $q$  is the incident photon momentum and  $a$  is the lattice constant of the material representing a typical range of bro-

ken inversion symmetry from which the nonlinear response is generated. The factor  $qa$ , however, is contained in the tensor  $\chi_{ijk}^{(2)}$  and has therefore not been written explicitly in Eq. (6) and all subsequent expressions for the nonlinear fields. This factor describes the ratio of linear excitation depth  $1/q$  and nonlinear response depth  $a$ .<sup>19</sup> The linear excitation depth  $1/q$  is connected to the usual electro-dynamical formula for the metallic skin depth

$$d_{\text{skin}} = \sqrt{\frac{1}{\mu_0 \sigma \omega}} = \frac{1}{q} \quad (\text{A2})$$

with conductivity  $\sigma$  and vacuum permeability  $\mu_0$ . This becomes for metals at frequencies below the plasma resonance  $\omega_p$

$$d_{\text{skin}} = \frac{c}{\omega_p}. \quad (\text{A3})$$

### APPENDIX B: REFLECTED SH FIELDS FOR ARBITRARY POLARIZATION ANGLE OF THE INCOMING LIGHT

In this appendix we give the general expressions resulting from the tensor  $\chi_{ijk}^{(2)}$  for the reflected  $p$ - and  $s$ -polarized SH field generated from  $\varphi$ -input polarization. In the longitudinal Kerr configuration one obtains for the reflected  $p$ -polarized SH field generated from  $\varphi$ -input polarization

$$E_{\varphi}^{(2\omega)}(p\text{-SH}) = 2i|E_0^{(\omega)}|^2 A_p [[F_c \chi_{xxz}^{(2)} 2f_c f_s + N^2 F_s (\chi_{zxx}^{(2)} f_c^2 + \chi_{zzz}^{(2)} f_s^2)] t_p^2 \cos^2 \varphi + (F_c \chi_{xxy}^{(2)} f_c + N^2 F_s \chi_{zyz}^{(2)} f_s) 2t_p t_s \cos \varphi \sin \varphi + N^2 F_s \chi_{zyy}^{(2)} t_s^2 \sin^2 \varphi] \quad (\text{B1})$$

and for the reflected  $s$ -polarized SH field generated from  $\varphi$ -input polarization

$$E_{\varphi}^{(2\omega)}(s\text{-SH}) = 2i|E_0^{(\omega)}|^2 A_s [(\chi_{yxx}^{(2)} f_c^2 + \chi_{yzz}^{(2)} f_s^2) t_p^2 \cos^2 \varphi + \chi_{yyz}^{(2)} 2f_s t_p t_s \cos \varphi \sin \varphi + \chi_{yyy}^{(2)} t_s^2 \sin^2 \varphi]. \quad (\text{B2})$$

In the polar Kerr configuration the tensor  $\chi_{ijk}^{(2)}$  yields for the reflected  $p$ -polarized SH field generated from  $\varphi$ -input polarization

$$E_{\varphi}^{(2\omega)}(p\text{-SH}) = 2i|E_0^{(\omega)}|^2 A_p [[F_c \chi_{xxz}^{(2)} 2f_c f_s + N^2 F_s (\chi_{zxx}^{(2)} f_c^2 + \chi_{zzz}^{(2)} f_s^2)] t_p^2 \cos^2 \varphi + N^2 F_s \chi_{zxx}^{(2)} t_s^2 \sin^2 \varphi + F_c \chi_{xyz}^{(2)} 2t_p t_s \cos \varphi \sin \varphi] \quad (\text{B3})$$

and for the  $s$ -polarized SH field

$$E_{\varphi}^{(2\omega)}(s\text{-SH}) = 2i|E_0^{(\omega)}|^2 A_s (\chi_{xxz}^{(2)} 2f_s t_p t_s \cos \varphi \sin \varphi + \chi_{xyz}^{(2)} 2f_c f_s t_p^2 \cos^2 \varphi). \quad (\text{B4})$$

### APPENDIX C: MAGNETIC PHASE SHIFT IN NONLINEAR OPTICS

In this appendix, we discuss the microscopic origin of the relative phase shift of  $90^\circ$  between the odd and even elements of the nonlinear susceptibility tensor  $\chi_{ijk}^{(2)}$ . First, we have to remark that this phase does not result from the fact that the nonlinear susceptibilities contain three matrix elements each yielding a factor of  $i$  rather than two in the linear case, since this difference occurs in the even as well as in the odd tensor elements. Instead, the microscopic origin is due to spin-orbit coupling which acts as a perturbation on one of the wave functions in the matrix elements of the odd tensor elements alone. For a plane-wave basis, for example, the spin-orbit perturbation yields the following identity,<sup>31,32</sup> which can be proven by commutator algebra

$$\langle \mathbf{k}' | \lambda_{s0} (\mathbf{k} \times \mathbf{s}) \nabla V | \mathbf{k} \rangle = i \lambda_{s0} V_{\mathbf{k}' - \mathbf{k}} (\mathbf{k} \times \mathbf{k}') \cdot \mathbf{s}, \quad (\text{C1})$$

thus giving a phase factor of  $i$  in the odd susceptibility tensor elements. This argument holds in the linear as well as in the

nonlinear case, but the resulting phase of  $i$  is compensated only in the linear case by the decomposition of  $\chi_{ij}^{(1)}$  yielding another factor of  $i$

$$\chi_{ij}^{(1)\pm} = \chi_{ij,0}^{(1)} \pm i \chi_{ij,1}^{(1)} \sin \theta. \quad (\text{C2})$$

This factor comes from the wave equation, which is homogeneous in linear optics. The susceptibility results directly from the dielectric function the square root of which is the eigenvalue of the wave equation, the complex index of refraction. The eigenmodes are left- or right-handed circularly polarized photons. In the nonlinear case, however, the decomposition has no factor of  $i$

$$\chi_{ijk}^{(2)\pm} = \chi_{ijk,0}^{(2)} \pm \chi_{ijk,1}^{(2)}, \quad (\text{C3})$$

since  $\chi_{ijk}^{(2)}$  is not related to the eigenvalues of the wave equation, which in nonlinear optics is an inhomogeneous differential equation.

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