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Nonlinear magneto-optics of freestanding Fe monolayers from first principles

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Abstract. The nonlinear magneto-optical Kerr-effect (NOLIMOKE) is determined for freestanding Fe monolayers with several in-plane structures from first principles. Based on the theory of nonlinear magneto-optics by Hübner and Bennemann [1] we calculate the nonlinear susceptibilities of the monolayers using the ab initio FLAPW-method WIEN95 with the additional implementation of spin–orbit coupling and the calculation of the dipole transition matrix elements appropriate for freestanding monolayers. We present results for the spectral dependence of the nonlinear susceptibility tensor elements and the resulting intensities and Kerr angles. Special emphasis is put on the effects of structural changes, such as the variation of the lattice constant and different surface orientations. The influence of spin–orbit coupling on the tensor elements for different magnetization directions is presented. Also, the azimuthal dependence of the intensities generated by several low-index surfaces is given, showing the pronounced sensitivity of second-harmonic generation to lateral structural changes and magnetic properties even in the monolayer range.

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The nonlinear magneto-optical Kerr effect (NOLIMOKE) combines the surface and interface sensitivity of second-harmonic generation (SHG) with the sensitivity to magnetic ordering, which is much stronger here than in linear optics [2, 3]. The existence and the detectability of the nonlinear magneto-optical effect was shown independently in 1989 by Pan, Wei and Shen [4] and Hübner and Bennemann [1]. Whereas the former work was based on a group-theoretical classification of the nonlinear susceptibility tensor $\chi^{(2)}_{ijk}$, the latter used an electronic structure calculation to predict the nonlinear Kerr effect.

During recent years significant improvements have been made. The group theoretical analysis was extended to antiferromagnetic surfaces [5]. Also several extensions of the electronic structure calculations have been performed, such as the calculation of the nonlinear magneto-optical response of Ni within a tight-binding scheme [6] and of Fe layers and films by using a FP-LMTO method [7]. In these papers, spin–orbit coupling (SOC) is treated perturbatively and only recently [8] was SOC included from first principles to obtain the nonlinear susceptibility of Fe monolayers for several in-plane structures. Nevertheless, no complete theory combining both the group theoretical analysis and first-principles electronic structure calculations has been available, since the dipole transition matrix elements had not been determined from first principles.

In this paper we present a calculation of the nonlinear magneto-optics for freestanding Fe monolayers, which obtains all fundamental aspects of magneto-optics, the electronic structure, including spin-polarization, SOC, and the dipole transition matrix elements from first principles. The symmetry aspects are completely determined by the symmetries of the wavefunctions.

1 Theory

Our calculations are based on the formula for the nonlinear susceptibility tensor,

$$\begin{aligned}\chi^{(2)}_{ijk}(2\boldsymbol{q},2\omega) = e^3 \sum_{\boldsymbol{k},l,l',l''} \Bigg\{ & \left\langle \boldsymbol{k}+2\boldsymbol{q}, l''|i|\boldsymbol{k}, l\right\rangle \left\langle \boldsymbol{k}, l|j|\boldsymbol{k}+\boldsymbol{q}, l'\right\rangle \\ & \times \left\langle \boldsymbol{k}+\boldsymbol{q}, l'|k|\boldsymbol{k}+2\boldsymbol{q}, l''\right\rangle \\ & \times \frac{\frac{f(E_{\boldsymbol{k}+2\boldsymbol{q},l''})-f(E_{\boldsymbol{k}+\boldsymbol{q},l'})}{E_{\boldsymbol{k}+2\boldsymbol{q},l''}-E_{\boldsymbol{k}+\boldsymbol{q},l'}-\hbar\omega+i\hbar\alpha_1} - \frac{f(E_{\boldsymbol{k}+\boldsymbol{q},l'})-f(E_{\boldsymbol{k},l})}{E_{\boldsymbol{k}+\boldsymbol{q},l'}-E_{\boldsymbol{k},l}-\hbar\omega+i\hbar\alpha_1}}{E_{\boldsymbol{k}+2\boldsymbol{q},l''}-E_{\boldsymbol{k}l}-2\hbar\omega+i2\hbar\alpha_1} \\ & \times \Bigg[1+4\pi\,\mathrm{e}^2 \sum_{ij} m_i m_j \sum_{\boldsymbol{k},l,l''} \left\langle \boldsymbol{k}l\,|\boldsymbol{r}_i|\,\boldsymbol{k}+2\boldsymbol{q}, l''\right\rangle \\ & \times \left\langle \boldsymbol{k}+2\boldsymbol{q}, l''\,\middle|\boldsymbol{r}_j\middle|\,\boldsymbol{k}, l\right\rangle \\ & \times \frac{f(E_{\boldsymbol{k}+2\boldsymbol{q},l''})-f(E_{\boldsymbol{k},l})}{E_{\boldsymbol{k}+2\boldsymbol{q},l''}-E_{\boldsymbol{k}l}-2\hbar\omega+2\mathrm{i}\hbar\alpha} \Bigg]^{-1} \Bigg\}, \end{aligned} \tag{1}$$

derived in [1]. Here $\boldsymbol{q}$ is the momentum of the incident light, α is a damping constant simulating a finite lifetime and chosen as 0.2 eV, $f(E_{\boldsymbol{k},l})$ denotes the Fermi-distribution

and m_i is the ith direction cosine of $\boldsymbol{q}$ with respect to the plane of incidence. The energy bands $E_{\boldsymbol{k},l}$ and the wavefunctions $|\boldsymbol{k}, l\rangle$ are determined by using the ab initio FP-LAPW method WIEN95 [9]. SOC is included as described in [8]. Within this treatment the wavefunctions reveal the correct symmetry of the system, including SOC. Thus, by determining the dipole transition matrix elements $\langle \boldsymbol{k}, l|i|\boldsymbol{k}', l'\rangle$ from the wavefunctions, we obtain the correct form of the susceptibility tensor with respect to symmetry, as derived in [4] from first principles. The numerical differences between group-theoretically allowed and forbidden tensor elements is about 12 orders of magnitude. The precise treatment will be described elsewhere [10].

Since, at this stage of the investigations, we are mainly interested in the effects of lateral structural changes on the magneto-optical spectra, we calculate only freestanding monolayers, neglecting the substrate. The absence of a substrate enables us to choose arbitrary in-plane structures and reduces the required computer time to a reasonable value. Of course, the substrate cannot be neglected for a quantitative determination of the magneto-optical response and will be included in further studies.

Within the electric-dipole approximation the presence of inversion symmetry breaking is necessary for the occurrence of SHG. Thus, in the case of freestanding monolayers we have to introduce an artificial symmetry breaking perpendicular to the surface. This is done by integrating the dipole transition matrix elements in the upper half of the unit cell only, which effectively sets the wavefunctions in the lower half volume equal to a constant. Roughly speaking, this simulates a substrate with a constant charge density. In doing so, we also solve the general problem of treating semi-infinite systems like film or layer structures with a $\boldsymbol{k}$-space method like WIEN95, which uses three-dimensional translational invariance.

The SHG intensities are then calculated from the nonlinear susceptibilities by using the expression for the second-harmonic-generated field given in [11].

2 Results

Figure 1 shows the spectral dependence of the imaginary part of the four independent tensor elements of the nonlinear susceptibility $\chi^{(2)}_{ijk}$ obtained from a Fe(001) monolayer with out-of-plane magnetization ($\boldsymbol{M} \parallel \boldsymbol{z}$) derived from a fcc lattice. The in-plane lattice constant is varied from 2.4 Å to 2.76 Å (Cu has $a = 2.56$ Å), simulating different substrates. For this system the nonlinear susceptibility tensor has the following form:

$$\chi^{(2)} = \left(\begin{array}{ccc|cc} 0 & 0 & 0 & xyz^- & xxz^+ & 0 \\ 0 & 0 & 0 & xxz^+ & yxz^- & 0 \\ zxx^+ & zxx^+ & zzz^+ & 0 & 0 & 0 \end{array}\right) . \qquad (2)$$

There are four independent tensor elements zxx^+, zzz^+, xxz^+ and xyz^-. The first three do not change their sign under magnetization reversal ($^+$ superscript) and thus will be called "nonmagnetic" [1], whereas the xyz element changes its sign

[1] Of course, these tensor elements also depend on the magnetization, but only in second order.

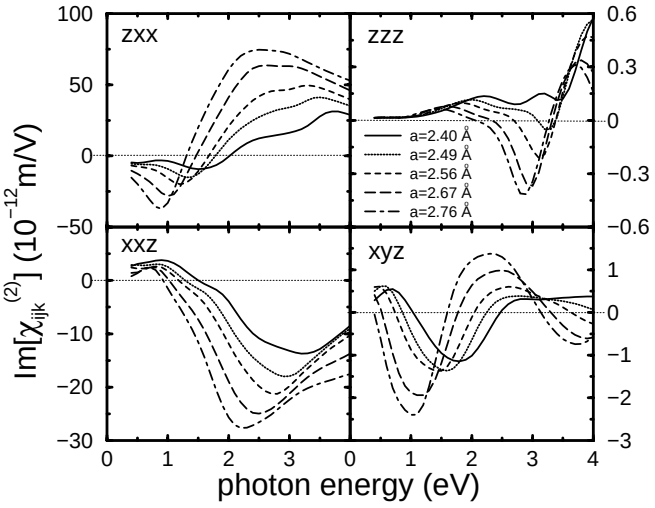

Fig. 1. Spectral dependence of the imaginary part of the nonlinear susceptibility tensor elements of a Fe(001) monolayer for different lattice constants a given in Å. The magnetization is out-of-plane ($\boldsymbol{M} \parallel \boldsymbol{z}$). The lattice constant is varied between 2.4 and 2.76 Å

($^-$ superscript) and is thus termed "magnetic". The tensor elements show a clear dependence on the lattice constant, as was already found in calculations with constant matrix elements [7, 8]. Zeros in the spectra are shifted to larger energies for smaller lattice constants; maxima have a larger magnitude for larger lattice constants. Of course, the spectral dependence becomes much more complicated with the inclusion of the matrix elements, since the latter affect both the width and absolute values of the resonances. The nonlinear Kerr angle $\Phi^{(2)}_{\mathrm{K,p}}$ in the polar configuration (p is the input polarization) is given by

$$\Phi^{(2)}_{\mathrm{K,p}} + \mathrm{i}\varepsilon^{(2)}_{\mathrm{K,p}} = \frac{A_s\, f_c\, f_s\, \chi^{(2)}_{xyz}}{A_p[N^2 F_s(\, f_c^2\, \chi^{(2)}_{zxx} + f_s^2\, \chi^{(2)}_{zzz}) + 2F_c\, f_c\, f_s\, \chi^{(2)}_{xxz}]} \qquad (3)$$

where $f_{s,c}$ are Fresnel coefficients of the incident field, $A_{p,s}$ and $F_{c,s}$ are transmission and Fresnel coefficients of the generated field and N is the refractive index of the substrate material. $\varepsilon^{(p)}_{\mathrm{K}}$ is the ellipticity, and an approximation for small angles is used. From Fig. 1 it is clear that the contribution of the $\chi^{(2)}_{zzz}$ and $\chi^{(2)}_{xxz}$ elements can be neglected because of their magnitude. Thus the nonlinear Kerr angle is directly proportional to the ratio of the $\chi^{(2)}_{xyz}$ and the $\chi^{(2)}_{zxx}$ elements weighted by the transmission and Fresnel coefficients (in the numerical calculation, however, all tensor elements are included). Though the spectral dependence of the Fresnel coefficients is rather weak in this energy range compared to the tensor elements, a clear relation between the spectral dependence of the Kerr angle and the tensor elements is not obvious. Nevertheless, both the spectral dependence of the Kerr angles and the relevant tensor elements for the different lattice constants (see Fig. 1) show some common features. Maxima around 1 eV are followed by a crossing of the curves between 1 and 1.5 eV. The spectral dependence of the linear Kerr angles in the upper part of Fig. 2 shows a similar feature at twice the energy but exhibits much more structure for higher photon energies. More detailed investigations of the complex quantities involved show that the spectral dependence of the nonlinear

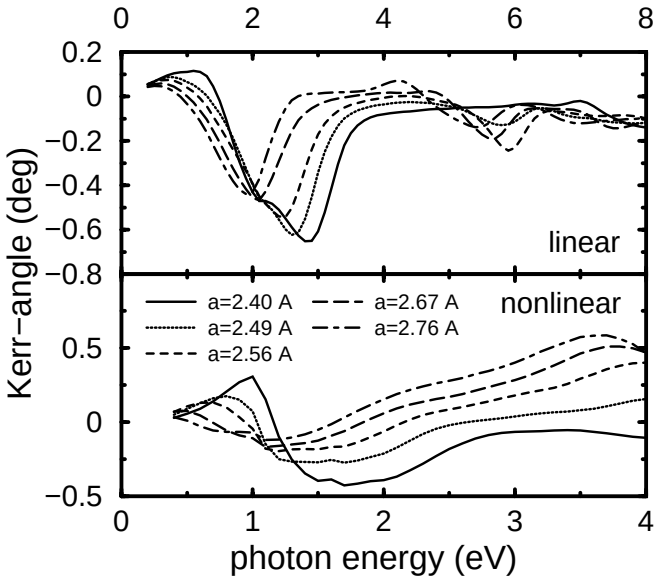

Fig. 2. Spectral dependence of the linear and nonlinear Kerr angle of a Fe(001) monolayer with out-of-plane magnetization. The lattice constant is varied between 2.4 and 2.76 Å

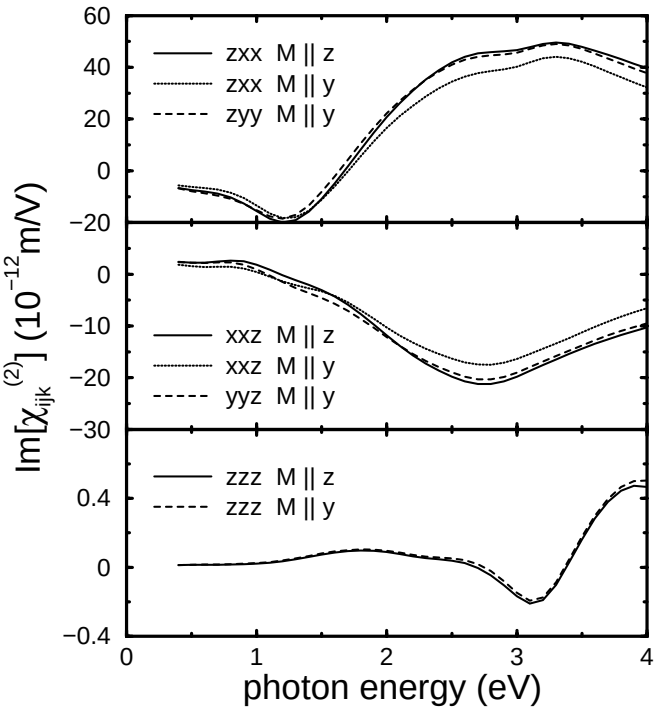

Fig. 3. Spectral dependence of the nonmagnetic tensor elements of a Fe(001) monolayer with out-of-plane and in-plane magnetization. The weak dependence reveals the SOC dependence of second order

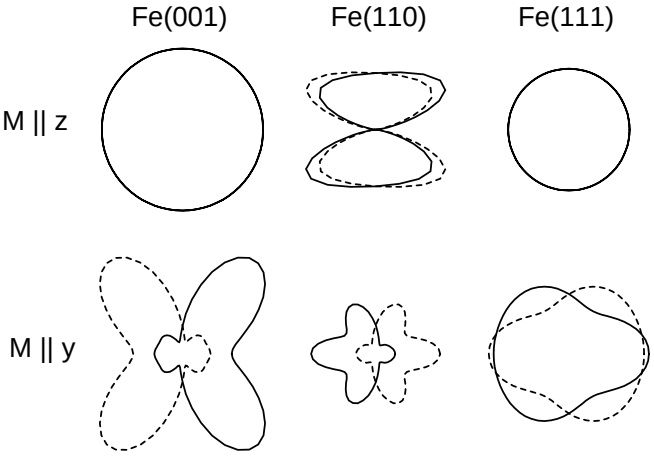

Fig. 4. Azimuthal dependence of the nonlinear intensities with p polarization of the incident and generated light for the Fe(001), Fe(110) and Fe(111) monolayer with out-of-plane and in-plane magnetization

Kerr angles is mainly ruled by the magnetic tensor elements. Figure 2 shows that in our calculations the linear and nonlinear Kerr angles are of the same order of magnitude, which does not agree with the experimental findings [12]. This is due to the specific form of the artificial inversion symmetry breaking performed here, which also underestimates the size of the *zzz* tensor element. Note, the present study focuses on the structural effects on the nonlinear Kerr spectra. This problem only occurs in the case of free-standing monolayers and can be overcome by including substrate effects and using unit cells for the electronic structure calculations in which inversion symmetry is explicitly broken [10]. Furthermore the size of the tensor elements in Fig. 1 is too large when compared with previous estimates [13]. This can be a problem of the neglected screening effects at the surface.

Figure 3 shows the differences in the spectral dependence of the nonmagnetic tensor elements of the Fe(001) monolayer with a lattice constant $a = 2.56$ Å for in-plane ($\boldsymbol{M} \parallel \boldsymbol{y}$) and out of plane ($\boldsymbol{M} \parallel \boldsymbol{z}$) magnetization. Since the two-dimensional unit cell of the (001) surface is a square lattice, the x and y directions are equivalent for out-of-plane magnetization, and thus $\chi^{(2)}_{zxx} = \chi^{(2)}_{zyy}$. For in-plane magnetization the x and y directions are no longer equivalent and thus $\chi^{(2)}_{zxx} \neq \chi^{(2)}_{zyy}$. The effect of this symmetry breaking on the nonmagnetic tensor elements is shown in Fig. 3. It shows only a slight dependence on the magnetization direction. This is expected from the fact that nonmagnetic tensor elements are only of second order in SOC [14].

One advantage of SHG over linear optics is the enhanced symmetry resolution resulting from using the third-rank tensor $\chi^{(2)}_{ijk}$ in SHG as opposed to the second-rank tensor $\chi^{(1)}_{ij}$ used in linear optics. The in-plane symmetry resolution can be monitored by calculating the azimuthal dependence of the intensities. This is displayed in Fig. 4, where the azimuthal dependence of the intensities with p_{in} and p_{out} polarization is plotted for the Fe(001), Fe(110), and Fe(111) monolayer with in-plane and out-of-plane magnetization. SHG can resolve up to three-fold rotational symmetries. Thus, for $\boldsymbol{M} \parallel \boldsymbol{z}$, the SHG signal of the square lattice Fe(001), which has four-fold symmetry in this configuration, shows no azimuthal dependence. Also the Fe(111) surface with $\boldsymbol{M} \parallel \boldsymbol{z}$ shows no azimuthal dependence since, in the absence of a substrate, this monolayer has a six-fold symmetry. The curve for the Fe(110) surface reveals its two-fold symmetry. The dashed line indicates the intensities for inverted magnetization direction, showing the effect of broken symmetries due to the magnetism and SOC. For an in-plane magnetization this effect is increased. The magnetization shifts the SHG intensities in the direction perpendicular to the magnetization. Also, the well-known effect of a large magnetic contrast in the intensities in the transverse

configuration is reproduced by our ab initio theory. Finally, it is clearly visible that the in-plane magnetization destroys the circular dependence of the intensities in the case of the Fe(001) and Fe(111) monolayers.

3 Summary and discussion

We have presented results on the nonlinear magneto-optical effect of Fe monolayers as obtained from first-principles calculations. The dipole transition matrix elements were computed from the wavefunctions. The inversion symmetry has been artificially broken to obtain SHG from the free-standing monolayers. The symmetries of the system including SOC are completely reflected by the wavefunctions yielding the correct form of the nonlinear susceptibility tensor known from group theoretical classifications [4]. The tensor elements and the resulting Kerr angle show a clear dependence on the lattice constant, in agreement with previous results from calculations using constant matrix elements. The spectral dependence of the nonlinear Kerr angle shows up as a complex superposition of the tensor elements and transmission and Fresnel coefficients involved. The precise inclusion of SOC reveals a weak dependence of the nonmagnetic tensor elements on the magnetization direction. The high sensitivity of SHG to the in-plane structure was shown for the Fe(001), Fe(110), and Fe(111) monolayers, also showing the important influence of the magnetic tensor elements.

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