Theory for Spin-Polarized Oscillations in Nonlinear Magneto-Optics due to Quantum Well States

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Abstract

Using an electronic tight-binding theory we calculate the nonlinear magneto-optical response from an x-Cu/1Fe/Cu(001) film as a function of frequency and Cu overlayer thickness (x=3...25). We find very strong spin-polarized quantum well oscillations in the nonlinear magneto-optical Kerr effect (NOLIMOKE). These are enhanced by the large density of Fe d states close to the Fermi level acting as intermediate states for frequency doubling. In good agreement with experiment we find two oscillation periods of 6-7 and 11 monolayers the latter being more pronounced.
The magnetism of low-dimensional metallic structures such as surfaces, thin films, and multilayer sandwiches has recently become an exciting new field of research and applications [1]. In particular, thin magnetic films and multilayers exhibit a rich variety of properties not previously found in bulk magnetism such as enhanced or reduced moments [2], oscillatory exchange coupling through nonmagnetic spacers [3–4], giant magnetoresistance [5], and the reorientation of the magnetic easy axis upon thickness and temperature variation [6–11]. Especially the observation of spin-polarized quantum well states (QWS) [12] in Cu/Co(001) has attracted a great deal of attention. It has become clear that quantum well states are indeed responsible for the important oscillatory behavior of the exchange coupling of ferromagnetic thin films via nonmagnetic spacers [16,17]. Presently mainly photoemission (PE) and inverse photoemission (IPE) [12–15] have been used to identify QWS effects. Very recently a possible connection between thickness dependent changes in NOLIMOKE and QWS [18] has been proposed.

It is the goal of this Letter to show that also nonlinear optics, in particular NOLIMOKE, is a new sensitive tool for studying QWS. We find very interesting structure in the NOLIMOKE signal due to particular transitions in \( k \)-space. This is very remarkable since it indicates that NOLIMOKE is able to detect very sensitively \( k \)-dependent structures. This new effect seems to be of general interest for the physics of nonlinear optics and its relationship to the underlying electronic structure. Note, this is not the case for linear optics, since there the contribution of the Drude term of the dielectric function creates a strong background of transitions from all \( k \)-directions. Nonlinear optics, in contrast to linear optics, is able to give angle-resolved information about the underlying electronic structure. We demonstrate this by extending previous work on the Fe/Cu(001) bilayer system [19] to the sandwich system \( x \)-Cu/Fe/Cu(001) where the layer number \( x \) is varied between 3 and 25. Thus we calculate the magnetic intensity contrast \( \Delta I_{2\omega} = \frac{I_{2\omega}(M) - I_{2\omega}(-M)}{I_{2\omega}(M) + I_{2\omega}(-M)} \) of NOLIMOKE for these systems and find very large quantum well oscillations, originating from particular transitions in \( k \)-space.

In view of the electronic structure presented in Fig. [1], a simple physical picture already
explains the occurrence of quantum well oscillations in NOLIMOKE. One gets the main
peak of the multilayer system, since for 11 layer and multiples of this the marked transitions
(a) between Cu $d$-bands and the quantum well states as final states become resonant at $2\hbar\omega$.
Obviously this causes an oscillation with a period of 11 monolayers (ML). Correspondingly
the period of 6-7 ML results from the marked transitions (b) in Fig. 1. Also it becomes
clear that the spin polarization of the intermediate Fe states will cause a magnetization
dependence and in particular a shifting of the peak for the magnetization direction $\mathbf{M}$ to
lower periods. For the situation sketched in Fig. 1 the $\mathbf{k}$-selectivity becomes immediately
obvious since unoccupied final states are necessary for a contribution to the SHG yield.

To verify these physical expectations we performed calculations using our previous the-
ory [20–22] to evaluate the SHG intensity $I_{2\omega}(\omega)$ for opposite magnetization directions. Em-
ploying an electronic theory for both the nonlinear susceptibility and the dielectric function
and separating $\chi^{(2)}(\omega)$ into even and odd parts under magnetization reversal $\chi^{(2)}_{\text{even}}(\omega)$ and
$\chi^{(2)}_{\text{odd}}(\omega)$, we get for the SHG yield within the electric dipole approximation for the polar
geometry (i.e. $\mathbf{M}$ normal to the surface) [23]

$$I_{2\omega}(\pm \mathbf{M}) = |2i| E_0^{(\omega)}|^2 \left\{ \chi^{(2)}_{\text{even}}(\omega) A_p [2 F_c f_c f_s \right.$$ 
$$+ N^2 F_s (f_c^2 + f_s^2) t_p^2 \cos^2 \varphi \cos \Phi] + \left\{ A_s [\chi^{(2)}_{\text{even}}(\omega) 2 f_s t_p t_s \cos \varphi \sin \varphi \right.$$ 
$$\pm \chi^{(2)}_{\text{odd}}(\omega) 2 f_c f_s f_p^2 \cos^2 \varphi] \} \sin \Phi \right|^2 \tag{1}$$

Here $\varphi$ and $\Phi$ denote the angle of polarization of the incident light and the outgoing second
harmonic light and were chosen as $\varphi = 0^\circ$ ($p$-polarized) and $\Phi = 75^\circ$. The linear amplitudes
$A_p$ and the transmission and Fresnel factors $t_{p,s}$, $f_{c,s}$ and $F_{c,s}$ are derived from the dielectric
function $\epsilon(\omega)$. Note, the nonlinear susceptibility tensor $\chi^{(2)}(\omega)$ is material specific via the
electronic bandstructure, and so are the linear dielectric function $\epsilon(\omega)$ and the indices of re-
fraction $n$ and $N$. To simplify our calculation, we assume constant matrix elements, which
are fitted to the linear dielectric function $\epsilon(\omega)$ [22,24]. This approximation is reasonable be-
cause the $\mathbf{k}$-dependence of the matrix elements is expected to become less important in two
dimensions due to the shrinking of the $d$-band width for the reduced coordination number and also due to the occurrence of additional allowed optical transitions \[22\]. Selection rules excluding dipole transitions of the type $\langle \Psi_{m=\pm 2}|r|\Psi_{m=\pm 0}\rangle$ were taken into account \[21\]. To compare with experiment \[24\]\[26\], we choose 1.61 eV as incident photon energy. A normalization with respect to the Cu layer number has to be performed to take the interface sensitivity of SHG into account in order to make the nonlinear response comparable for various film thicknesses. Therefore, we divided $I_{2\omega}(\omega)$ by the layer number, ensuring that for a band structure without dispersion the response is identical for all layer numbers. To calculate $\epsilon(\omega)$ and $\chi^{(2)}(\omega)$ from the electronic band structure of the x-Cu/1Fe/(001)Cu system we use a Cu bulk Hamiltonian (thus depending on $k_x$, $k_y$, $k_z$; $k_z = k^\perp$ is perpendicular to the layers ) combined with a Fe monolayer. The Hamiltonian is calculated within the Combined Interpolation Scheme \[27\], the parametrisation is according to Fletcher and Wohlfahrt \[28\]. The parameters for the Cu bulk bandstructure are taken from \[29\], for the Fe monolayer they have been achieved from a fit to an ab initio calculation \[30\]. Of course, in $\Gamma$ - X direction there is no dispersion of the Fe monolayer band structure. We are evaluating the SHG response at $(k_x, k_y) = (0,0)$, since for the (001) direction the high density of states due to the extremal Fermi surface diameter (caliper) at $k_\parallel = (0,0)$, which give the QW period from Ruderman-Kittel-Kasuya-Yoshida (RKKY) calculations \[31\] dominates the output \[32\]\[33\]. The $k$-summation is performed over $k$-points along the $k^\perp$ direction.

Note, due to the two resonance denominators of $\chi^{(2)}(\omega)$ \[24\] it is not necessary for the intermediate state to be unoccupied to give a contribution to $\chi^{(2)}(\omega)$. It is sufficient if the final state (usually a Cu $s$ state) is unoccupied. As a consequence, a high density of intermediate states leads to a large number of contributing terms to $\chi^{(2)}(\omega)$, thus enhancing the SHG response. In our electronic structure, this amplification is caused by the spin polarized Fe $d$ states. Since at least one of the three states involved in a nonlinear transition must be unoccupied to give a contribution to the SHG yield, the QWS above $E_F$ are of great importance as final states for the NOLIMOKE signal. The QWS result from the confinement of the electrons in thin films, causing an equally spaced discretization in $k^\perp$-direction,
whereby the number of \(k\)-points equals the number of layers. Clearly this discretization of the \(k\)-values affects the SHG intensity since photon transitions are limited to these distinct \(k^\perp\) points.

For the fundamental period \(\Lambda\) of intensity oscillations as observed in photoemission, transitions at \(k^\perp\)-vectors are decisive, for which (for Cu bulk) the \(s\)-band crosses the Fermi surface. If the layer number increases, such unoccupied \(k^\perp\)-states at the Fermi surface occur if the layer number equals \(m \cdot k^\perp_{BZ}/(k^\perp_{BZ} - k^\perp_F)\), with \(m = 1, 2, 3, \ldots\). Then these new unoccupied \(s\)-states permit additional transitions, and the optical response increases. The ratio \(k^\perp_{BZ}/(k^\perp_{BZ} - k^\perp_F)\) gives the fundamental period \(\Lambda\). Obviously, since optical transitions may occur to all states above \(E_F\), this period only marks a lower limit of possible oscillation periods and is not as strict as for (I)PE experiments, and depends on the photon energy and the position of the initial bands. In particular for the nonlinear response, due to the additional degree of freedom and due to \(2\hbar\omega\) resonances the SHG intensity increases and new (larger) oscillation periods occur. Although every period longer than the fundamental one may occur in the SHG spectrum if \(d\) states allow for resonances with unoccupied QWS at a \(k^\perp\) vector between \(k^\perp_F\) and \(k^\perp_{BZ}\), the period \(\Lambda\) obtained from photoemission experiments and the doubled period \(2\Lambda\) have an outstanding importance. If a QWS allows for a SHG signal with period \(2\Lambda\), there is a QWS at \(k_F\) too, both resonant transitions (a) and (b) indicated in Fig. 1 contribute to the SHG signal at layer thickness \(n \cdot 2\Lambda\), thus enlarging the SHG amplitude at \(m \cdot 2\Lambda\). Due to interferences of the various transitions, this enhancement is not compensated by the performed normalization. This effect is only present for multiples of \(2\Lambda\) and is completely absent in linear optics. Our calculation shows that the spin polarized Fe \(d\) bands are responsible for the occurrence and amplification of the observed oscillations. This becomes apparent if we compare the resulting SHG intensity of the x-Cu/1Fe/Cu(001) system for opposite magnetization directions with the system without Fe interlayer (but keeping the confinement for the Cu overlayer), which is 50 times weaker, in good agreement with experiment [25]. This strong enhancement is caused by the additional terms to be summed for the calculation of \(\chi^{(2)}(\omega)\) when more bands are present, even if they are not
resonant with the QWS. This amplification mechanism is not possible in linear optics, in agreement with experimental observations [34].

In Fig. 2 we show results of our calculation of the NOLIMOKE signal demonstrating the pronounced QWS oscillations and their strong spin dependence. The large peak at approx. 11 ML (and a corresponding peak at 22 ML) results from the amplification due to the Fe bands, while the resonant $2\hbar \omega$ transition is between Cu $d$ and Cu $s$ bands (transition (a) in Fig. 1). Since the position of the Fe bands is less important for such a constellation, this peak is dominating the SHG spectrum for both magnetization directions. At 6-7 ML the Cu $d$ band edge is too far below $E_F$ to give resonances with the QWS for the photon energy of 1.61 eV, hence a much reduced intensity results. While for the majority spins both $\hbar \omega$ resonances with Fe $d$ bands as intermediate states and $2\hbar \omega$ resonances with Fe as initial state are important, the minority transitions involve mainly $\hbar \omega$ resonances with intermediate Fe $d$ states and Cu $d$ states as initial states. Since these resonances are not well matched by the photon energy, the short period of the SHG yield from the minority electrons is less pronounced. This can be traced back to the $I_{2\omega}(-M)$ yield, which is (in the geometry under consideration) influenced mainly by the minority transitions. The observed slight difference of the corresponding periods between the two magnetization directions is caused by the exchange splitting of the Fe $d$ bands, allowing for resonances with the QWS at different layer numbers. The inset of Fig. 2 showing results for the magnetic contrast $\Delta I_{2\omega}$ gives further evidence for the importance of the Fe $d$ bands. The result indicates clearly that the exchange splitting of the Fe interlayer is involved. The contrast varies between 100% and $-80\%$ and changes sign several times, due to the same magnitude of the SHG intensity for both magnetization directions. This coincidence of the two intensities at fixed frequency could not be explained if the SHG yield would be generated solely by transitions between three spin polarized quantum well states, since then one signal should be much more pronounced than the other one. Furthermore, then the signal of a pure Cu surface should be of the same magnitude as that of the sandwich. Calculations for different exchange splittings showed that the occurring oscillation periods and the phase shift between the SHG
yield for opposite magnetization direction are strongly influenced by the strength of the spin splitting. These results indicate a suppression of periods at particular exchange energies in sandwich structures.

In Fig. 3 we show the dielectric function $\epsilon(\omega)$, its $M$-dependence and the linear Kerr angle. These results demonstrate the enhanced sensitivity of NOLIMOKE regarding oscillations due to QWS as compared with the linear optical response. Two oscillation periods for the imaginary part of the dielectric function $\epsilon(\omega)$ for both magnetization directions can be seen, a dominant one with period 7 ML and a less pronounced oscillation with a period of 3-4 ML. From Fig. 1 the origin of these oscillation periods becomes clear, since for about 6-7 ML (and multiples of these thicknesses) there are QWS above $E_F$ resonant with a Fe minority $d$ Band. Similarly, for the other peaks there are respective resonances with Fe $d$ bands. In contrast to nonlinear optics, an overall increase of the linear signal with Cu thickness is observed, since it results not only from the interface, but from all layers, so that the normalization with respect to the layer thickness has not to be performed. Of course, the doubled period (11 ML) is absent, since $2\hbar\omega$ resonances do not contribute to the linear signal. Note, the magnetic effect is three orders of magnitude smaller than for the nonlinear signal, due to the strong influence of the nonmagnetic intraband transitions on the linear signal. Thus for the linear susceptibility, $\chi_{\text{even}}^{(1)}(\omega, M) \gg \chi_{\text{odd}}^{(1)}(\omega, M)$. This small magnetic effect becomes obvious from the linear Kerr angle $\Phi_{\text{Kerr}}$ shown in the inset of Fig. 3. $\Phi_{\text{Kerr}}$ is of the order of mdeg, whereas the nonlinear Kerr angle is two to three orders of magnitude larger. The overall increase with increasing layer thickness is again due to the long range of MOKE. The period of 6-7 ML is due to $\hbar\omega$ resonances between QWS above $E_F$ and the Fe majority $d$ band at -1.4 eV. If this transition is resonant, the majority contribution of $\epsilon(\omega)$ increases, resulting in an increase of $\Phi_{\text{Kerr}}$ at the corresponding layer thickness.

In conclusion, we showed that QWS give rise to strongly enhanced oscillations. The electronic origin of this strong enhancement is analyzed. We get that NOLIMOKE is able to probe particular transitions in $k$-space. Our results demonstrate that although caused by the $s$ QWS the amplitude of the oscillation is due to the high density of Fe $d$ states. Periods
different from the fundamental period found in PE experiments are possible, depending on
the position of resonant $d$ bands below $E_F$. In contrast to linear optics, in NOLIMOKE even
$2\hbar\omega$ resonances strongly influence the oscillation. In the considered sandwich structure, this
makes the doubled period to dominate the spectrum.

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It remains to be shown that contributions which in principle could give rise to additional oscillations play a significant role.
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FIGURES

FIG. 1. Band structure of the xCu/1Fe/Cu(001) sandwich along $k^\perp$. Bands with energy less than -5 eV are not drawn. The numbers 5, 6, 8, 10, 12 on the Fermi energy level indicate the $k$-position of unoccupied QWS as they occur at the corresponding layer number. The marked transition (a) has a resonance at $2\hbar\omega$ and is responsible for the main peak at approx. 11 ML. The nonlinear transition (b) is responsible for the oscillation period of 6-7 ML. Also the dominating transition for MOKE making the 6-7 ML oscillation is indicated (dashed arrow).

FIG. 2. SHG yield for opposite magnetization directions $\mathbf{M}$ and $-\mathbf{M}$. The dominating 11 ML period is due to a $2\hbar\omega$ resonance between Cu $d$ states and quantum well states, drastically enhanced by the Fe $d$ bands and thus demonstrating the $k$-selectivity of NOLIMOKE. The signal for neglected Fe bands is nearly vanishing on this intensity scale. The peak shift between the $\mathbf{M}$- and $(-\mathbf{M})$-signal is due to the spin polarization of the Fe $d$ bands. $I_{2\omega}$ refers to the case where Fe is absent, but the confinement of the Cu layers is kept. The inset shows the magnetic contrast $\Delta I_{2\omega} = \frac{I_{2\omega}(\mathbf{M})-I_{2\omega}(-\mathbf{M})}{I_{2\omega}(\mathbf{M})+I_{2\omega}(-\mathbf{M})}$.

FIG. 3. Linear dielectric function of the x-Cu/1Fe/Cu(001) sandwich for opposite magnetization as a function of the Cu layer thickness. Note, the 6-7 ML period is visible, while the 11 ML period is completely absent. The magnetic contrast is much smaller than for the NOLIMOKE signal. The inset shows the linear Kerr angle as a function of the Cu layer thickness.
