Thickness-dependent oscillation of the magneto-optical properties of Au-sandwiched (001) Fe films

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Au-sandwiched bcc (001) Fe wedges were produced by the molecular beam epitaxy method (0–20 Å). The polar Kerr rotation and Kerr ellipticity spectra (1.5–5 eV) were measured as a function of the Fe thickness d. In both kinds of spectra new peaks appear. The complex Kerr rotation oscillates as a function of the film thickness. More than four oscillations could be clearly observed in the measured thickness range. Magneto-optical transitions from the Δ_5 band to quantum-well states (QWS's) in the Δ_1 majority-spin band change the intrinsic dielectric properties. The oscillation period and transition energies are in agreement with theory. Thickness-dependent structures below 2.5 eV strongly suggest the occurrence of partially confined QWS's around E_F or QWS's in the Δ_5 band.

I. INTRODUCTION

Much research has been done on magnetic multilayer systems. Special properties such as a large perpendicular anisotropy, an enhanced Kerr rotation at short wavelength, a giant magnetoresistance (GMR) effect, or an oscillation of the exchange coupling make them very interesting for applying in sensors, communication devices, or recording media. Furthermore study of multilayers is also interesting from a physical point of view; the study of metallic thin films by photoemission spectroscopy and inverse photoemission spectroscopy (IPES) showed the existence of a discrete density of states.[1,2] These so-called quantum-well states (QWS's) play an important role in the exchange coupling[3] between two ferromagnetic layers. From the oscillating character[4] of this coupling information, the Fermi surface of the nonferromagnetic intermediate layer can be obtained. However the role of the ferromagnetic layer[5,6] in the coupling process is not completely clear at the moment. From the theoretical approaches of Barnas[7] and Bruno,[8] an oscillation of the exchange interaction as a function of the ferromagnetic film thickness is predicted. In this case the period is determined by the stationary spanning k vector on the Fermi surface of the ferromagnetic material.

More recently, it was shown that the intrinsic magneto-optical (MO) spectra of Au-sandwiched ultrathin Fe films[9] strongly deviate from the bulk values.[10] A new peak appeared to exist which shifts to higher energies for thicker film thicknesses. In order to observe the new transition, apparently very smooth films (on an atomic scale) have to be used.[11] By using the theoretical band structures of Au and the IPES data of Fe ultrathin films,[2] this peak could be interpreted as the result of a transition from the $\Delta_5\uparrow$ band to a QWS in the $\Delta_1\uparrow$ majority-spin band.[11]

In order to obtain a better understanding of quantum-well states in thin Fe films and their possible influence on the coupling process, we performed a systematic study on the magneto-optical finite-size effects of monocrystalline ultrathin (001) Fe films. The experiments were performed on wedge samples, which guarantees a large accuracy of the relative film thickness. In addition to that an improved new MO spectrometer was used. Both Kerr rotation and Kerr ellipticity were directly measured without the use of the Kramers-Kronig relations. This made an accurate study of the thickness dependency of the complex rotation possible.

II. EXPERIMENT

The samples (see Fig. 1) were prepared by the molecular beam epitaxy (MBE) deposition technique in a chamber with a background pressure in the 10^{-10} Torr range. Polished or in-air-cleaved MgO (100) samples were used as a substrate. Before deposition the substrate was heated to 950 °C for 1 min in order to remove adsorption gases and stress introduced by the cleaving or polishing process. After cooling down till room temperature a Ag buffer layer of 2000 Å was deposited. This Ag layer

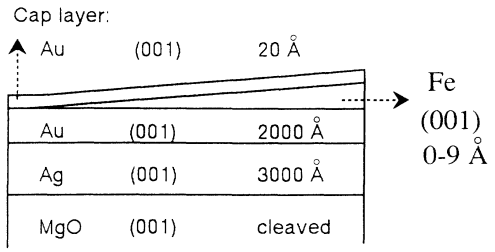

FIG. 1. Schematic cross section of the Au-sandwiched Fe wedge (sample B): (a) Au(20 Å)/Fe(0.1 Å/mm)/Au(2000 Å)/Ag(2000 Å) on cleaved MgO(100): 0–21 Å (sample C); (b) Au(20 Å)/Fe(0.05 Å/mm)/Au(2000 Å)/Ag(2000 Å) on cleaved MgO(100): 0–10 Å (sample B); (c) Au(20 Å)/Fe(0.05 Å/mm)/Au(20 Å)/Ag(3000 Å) on polished MgO(100): 0–10 Å (sample A).

was smoothed by annealing the sample for 1 min at 450 °C. On top of the Ag base a 2000 Å seedlayer of Au was deposited. This layer was smoothed by an annealing step of 300 °C. As shown by others[12] epitaxially layer by layer growth of Fe can be obtained on a Au (001) substrate. In order to avoid segregation of Au (Ref. 13), on top of the deposited Fe, and in order to guarantee sharp interfaces, the deposition of the Fe layer was done at room temperature.[14] The Fe wedge was created by moving the sample under a shutter during deposition. Two types of wedges were prepared: steep wedges with a slope of 1 Å/mm (sample C) and normal wedges with a slope of 0.5 Å/mm (samples A and B). During deposition the growth mode was monitored by reflection high-energy electron-diffraction (RHEED) experiments. Clear oscillation of the intensity can be observed for this growth mode (Frank van der Merwe growth). In order to prevent oxidation the multilayer structure was covered with a Au caplayer of 20 Å. After deposition of 10 Å of Au the 1×5 RHEED reconstruction pattern could already be clearly observed. The crystal structure was checked by x-ray analysis. A schematic cross section of the samples is shown in Fig. 1.

The MO experiments were performed with an improved Jasco-2500 spectrometer. This spectrometer is based on the piezobirefringent modulator technique.[15] By choosing long integration times large accuracy could be obtained. All measurements were done in the polar configuration (field perpendicular to the film surface, angle of incidence 10° from the film normal).

The an-hysteresis curves (this is a hysteresis curve measured with an ac magnetic field superposed on the normal dc magnetic field) were determined as a function of the film thickness by measuring the ellipticity as a function of the applied field (ac field 200 Oe, 0.2 Hz, maximum dc field 16 kOe).

The spectra were measured in the photon energy range of 1.5 to 5 eV. A maximum field of 16 kOe was used. Because of the aid of the surface anisotropy[16] this was enough to saturate the sample positions with an Fe thickness of less than 8 monolayers (ML). For the positions with thicker Fe layers a correction had to be made. The correction was estimated from extrapolating the relation between the saturation field and the film thickness. The bandwidth of the monochromator was kept 2 nm over the complete measurement range. Measurements performed with a constant slit width gave similar results.

Besides the spectra, so-called position scans were measured; the Kerr rotation and Kerr ellipticity were determined as a function of the film thickness. By choosing integration times of over 2 min and alternating the field, a high accuracy was obtained. All position scans were performed with a bandwidth of 2 nm. The spot size varied from 0.3 mm in the low-energy range to 1.5 mm for 5 eV. From this size and the discrete character of the interface, the Fe thickness bandwidth of the here-presented results is estimated to be smaller than 0.7–1 ML. All MO measurements were performed at room temperature in open air. On similar samples no change in MO properties could be observed over a time span of more than one year.

III. MEASUREMENT RESULTS

Figure 2 shows the saturation field (H_S) as determined from the measured ac-hysteresis curves as a function of the Fe film thickness. Within the measurement error and measurement interval, the behavior can be considered to be almost linear. From this we conclude that the Kerr rotation of the films with an H_S larger than 16 kOe can be estimated by using a correction factor determined from extrapolating this curve.

Typical polar Kerr rotation spectra for different Fe thicknesses (sample B) are presented in Fig. 3(a). For comparison the measurement values are normalized to the film thickness. Besides the large peak around 2.5 eV that is caused by the plasma edge of the Au substrate,[17,18] several new thin-film peaks were found. New structures can be clearly observed at 3, 3.5, 4, and 4.5 eV. In Fig. 3(b) the polar Kerr ellipticity spectra are shown. The same peaks can be observed as in the Kerr rotation spectra. Because of the larger signal-to-noise ratio much more detail could be observed in the Kerr ellipticity data. A thickness-dependent structure around 2.25 eV can be clearly observed.

Figure 4 shows position scans of the Kerr ellipticity for

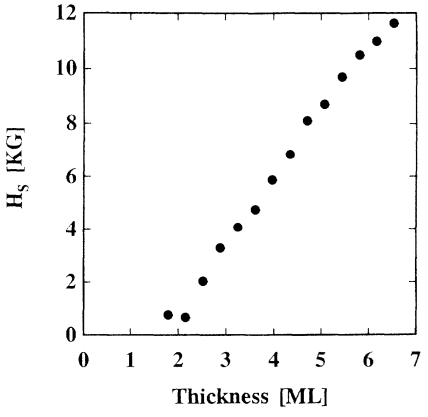

FIG. 2. Saturation field, H_S, as a function of the film thickness.

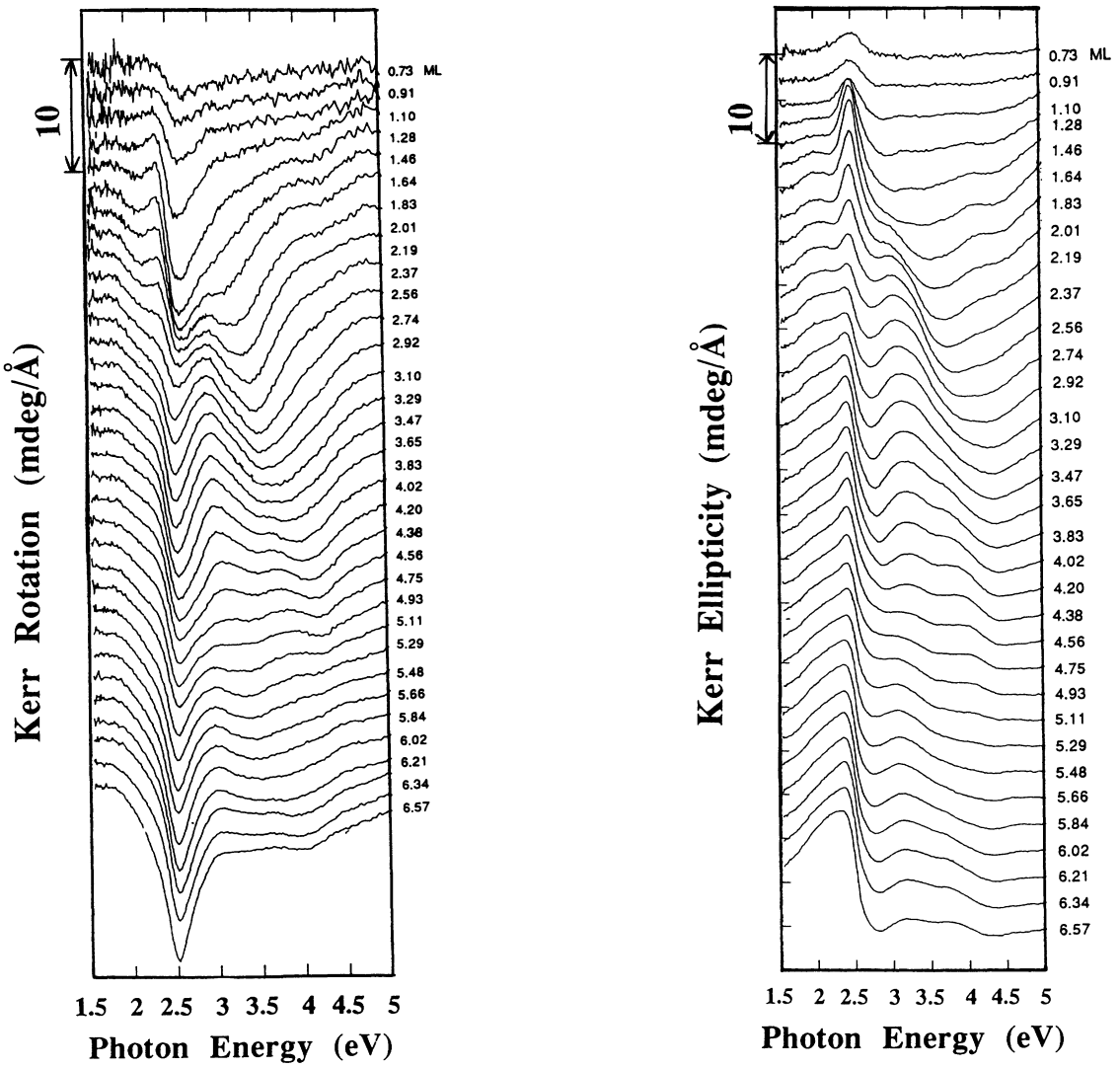

FIG. 3. Magneto-optical spectra (sample B) for different Fe thicknesses normalized to a unit film thickness: (a) Kerr rotation and (b) Kerr ellipticity.

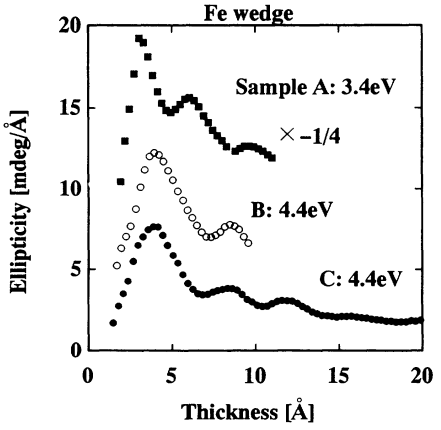

FIG. 4. Oscillation of the Kerr ellipticity as a function of the film thickness for sample A (3.4 eV), B (4.4 eV), and C (4.4 eV); The curve of sample A is multiplied by −0.25 for fitting in the plot.

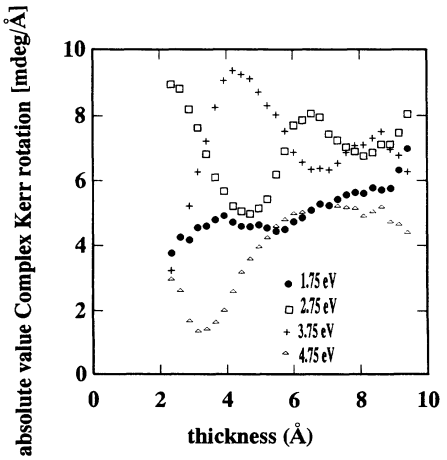

FIG. 5. Oscillation of the absolute value of the normalized complex Kerr rotation for several photon energies (sample B).

three different films. Clear oscillations appear to exist for all the samples. Scans performed with a smaller field amplitude showed the same behavior.

In Fig. 5 we plotted the absolute value of the normalized complex Kerr rotation (in [mdeg/Å]) as a function of the film thickness for sample B. The periods as determined from the plots appear to be in the range of 2.6–3.3 ML. The period seemed to increase for larger photon energies. However more careful analyses have to be performed. Largest amplitudes are observed for photon energies between 2.5 and 4eV; values to about 30% of the average rotation were measured.

IV. DISCUSSION

From the behavior of the saturation field as a function of the film thickness we conclude that the Kerr oscillations cannot be explained by an oscillation of the anisotropy constant. It is clear that films thinner than 8 ML can be easily saturated by a field of 15 kOe. The oscillations of Figs. 4 and 5 also cannot be understood by considering optical interference effects in the thin Fe film. For interference effects the effective optical path length must vary by $\lambda/2$ in the Fe thin film. Regarding the optical constants of bulk Fe this should lead to an oscillation period of about 1000 Å, much larger than the one found in this work. Furthermore in the thickness range below 100 Å, the theoretical Kerr rotation as calculated from the Maxwell equations, will be almost linear as a function of the film thickness[17–20] and will certainly not show any oscillations. It can be easily shown that the relation between the complex Kerr rotation and the dielectric properties of substrate and thin film is given by[9]

$$\phi_k + i\eta_k = i\frac{4\pi d}{\lambda}\frac{\varepsilon_{xy}}{1-\varepsilon_{xx}^s}\,, \tag{1}$$

where $\phi_k + i\eta_k$ is the complex Kerr rotation, d is the film thickness, λ is the wavelength, ε_{xy} is the off-diagonal component of the dielectric tensor of the Fe thin film, and ε_{xx}^s is the diagonal component of the dielectric tensor of the substrate. This approximation is valid for $2\pi d/\lambda \ll 1$.

The new peaks and observed oscillations as described above are apparently due to an oscillation of the off-diagonal component of the intrinsic dielectric tensor of the thin Fe film. The period of about 2.8 ML rejects the idea that the oscillations are caused by a periodicity of the interface roughness between the Fe layer and the Au caplayer. The oscillation of the Fe roughness, monitored by RHEED-oscillation experiments during the film growth, has a periodicity of 1 ML. From effective-medium approximations,[21] it is clear that a surface roughness could change the off-diagonal components of the dielectric tensor. In our measurement results however no oscillation with a period of 1 ML could be detected.

In Fig. 6 part of the bulk band structures of bcc Fe (Ref. 22) and fcc Au (Ref. 23) is shown. For the electrons belonging to the $\Delta_1\uparrow$ band of Fe, there are no states available in the Au between 1.7 and 6.3 eV above the Fermi level. For this reason their electron wavefunctions will be reflected at the Fe/Au interfaces and they will be confined to the Fe layer; the $\Delta_1\uparrow$ band will be discretized in the above-mentioned energy range.[24] Especially in the thinner Fe range it is expected that because of the small number of QWS's the spectra will be largely different from its bulk shape. The shift of the QWS's to higher energy for thicker Fe films will lead to a thickness-dependent oscillation of the Kerr rotation. Although the relation between QWS's and Kerr spectra is much more complicated than that between QWS's and the exchange coupling through a noble metal,[25] the oscillation period can be estimated from the dispersion of the $\Delta_1\uparrow$ band. Assuming that the QWS's in the Δ_1 band are most important for the magneto-optical finite-size effects we estimated the oscillation period from the k vector at 1.7–4 eV above the Fermi level. The following equation, similar to, for example, in Ref. 2 was used:

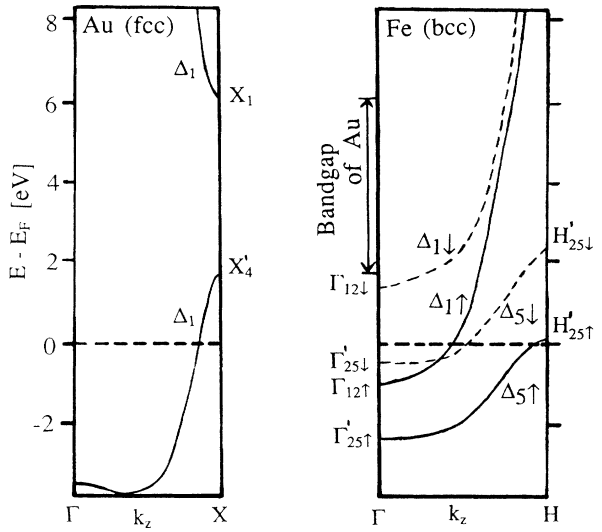

FIG. 6. Bulk band structures of Fe and Au around the Fermi level.

$$P(E) = \frac{k_{zb}}{k_{zb} - k_E}\,, \tag{2}$$

where $P(E)$ is the period of occurrence of a QWS at an energy E eV above the Fermi level; k_{zb} is the wave vector of the Brillouin-zone boundary; k_E is the wave vector E eV above the Fermi level. k_E was estimated from the theoretical band structure of Ref. 22.

The calculated value of the period varies from 2.6 to 3.4ML for larger E. The trend is in agreement with the experimental results of Fig. 5. These calculated periods are different from the period of the exchange oscillation as determined by Okuno and Inomata[6] for Fe(d Å)/Cr/Fe(d Å) (100) multilayers. They found that the coupling constant oscillates with a period of 5.6 ML as a function of the Fe thickness; for the oscillation of the exchange coupling the period is determined by the stationary spanning k vectors on the Fermi surface. However for the MO spectra we have to take account of the densi-

ty of states in a much more wide energy interval around E_F.

As shown above by Eq. (1), the positions of the peaks in the ϕ_k spectra are influenced by the optical properties of the substrate. However the positions of the maxima/minima in the plots of Fig. 5 are not influenced by the optical properties of the substrate. This can be easily shown by taking the absolute value on both sides of Eq. (1) and rearranging:

$$|\varepsilon_{xy}| = \frac{\lambda}{4\pi} |1-\varepsilon_{xx}^{s}| \frac{|\phi_k + \eta_k i|}{d} . \quad (3)$$

Since for one wavelength value the multiplication factor between the quantities $|\phi_k + \eta_k i|/d$ and $|\varepsilon_{xy}|$ is constant, a peak in the graph of $|\varepsilon_{xy}|$ as a function of the film thickness coincides with a peak in the graph of $|\phi_k + \eta_k i|/d$ as a function of the film thickness (this is also the reason why we plotted the normalized rotation in Fig. 5). Assuming that the new transitions will not overlap (narrow peaks in the spectra), we can obtain exact transition energies from the plot of Fig. 5 without having to know the dielectric properties of the substrate. It should be stressed here that it is not possible to determine the transition energies directly from the spectra because the position of the peaks is influenced by the optical properties of the substrate.

In order to minimize the influence of a possible background curve, we calculated the transition energies from the maxima of the second derivative of Fig. 5. The results are presented by the black dots in Fig. 7 and will be related to the results of a very simple nearly free-electron (NFE) approximation calculation in the section below. The background curves in Fig. 5 may be caused by the change of Curie temperature,[12,14] caused by the reduction of the dimensionality of the thin film.

Although complete band calculations have to be performed in order to calculate the MO spectra of this thin-film system, the influence of the finite dimension of the thin film on the spectra can be estimated by performing a one-dimensional NFE-based calculation of Δ_1 QWS's using the parameters of the theoretical band structures of bulk Fe and bulk Au.[22,23]

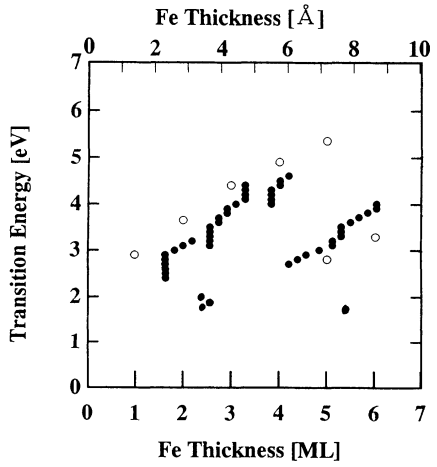

FIG. 7. Calculated (circles) and measured MO transition energies (black dots) versus film thickness.

We assumed the electron potential in Fe to be constant and in Au to be sinuslike with a period equal to half the lattice parameter and an amplitude equal to half the bandgap of the Δ_1 band (gap between $X4'$ and $X1$ points). We did not consider the p-d hybridization of the Δ_1 band in Fe; the offset between the electron potential of Fe and Au is approximated by the difference between the Γ_1 point of Fe and the Γ_1 point of Au. From the calculated wave functions and the quantization condition, the energies of the QWS at the $\bar{\Gamma}$ point in the two-dimensional Brillouin zone were calculated (we did not consider the redistribution of the electronic charges due to the difference in the work function of Au and Fe (0.8 eV) (Ref. 26) and we did not try to model the interface electronic structure).

The optical transitions from the $\Delta_5\uparrow$ to the $\Delta_1\uparrow$ quantum-well states are considered to contribute to the magneto-optical size effect in the thin-film range. The results of this calculation are presented by the open circles in Fig. 7. The same tendency is found for experimental and theoretical data for the quantum numbers $n=1$ and $n=2$. The black dots at low energies, which coincide with the structures in the MO spectra around 2.25 eV, were checked by measuring accurate position scans on sample C (bandwidth 10 nm). A weak oscillation of at least two periods was found in the Kerr rotation scan of 2.25 eV. For lower energies the oscillations' character strongly decreases (sample C). The low-energy data of Fig. 7 cannot be explained by discrete QWS's in the $\Delta_1\uparrow$ band. However, because of partial reflection of the electrons at the Fe/Au interface, partially confined QWS's may appear in the $\Delta_1\uparrow$ band of Fe below the x_4' point of Au. The large difference in the k vector between Fe and Au near the Fermi level suggests a nonzero electron reflection coefficient at the Fe/Au interface which implicates partial confinement of the electrons. Although this will not lead to a discretization of the energy levels, it may modulate the density of states. Simple calculations show enhancements of the density of states in the $\Delta_1\uparrow$ band of Fe. Another possibility for the observed oscillations at low photon energy is the occurrence of QWS's in the Δ_5 band. D-like QWS's have been observed in Pd and Au noble-metal overlayers on Fe and Co,[27] but not yet for thin Fe films. Further investigations on partially confined QWS's and d-like QWS's are necessary. In addition to that a theory that describes the MO oscillations in terms of QWS's and partially confined QWS's should be developed.

V. CONCLUSIONS

The above presented measurement results clearly illustrate the thickness dependence of the intrinsic dielectric properties of ultrathin Fe films. From the appearance of new peaks in the MO spectra and the oscillation of the absolute Kerr rotation as a function of the film thickness, we conclude that these thin-film MO transitions are caused by finite-size effects. A simple calculation shows

that the trend in the measurement results can be explained by MO transitions from the $\Delta_5\uparrow$ band to QWS's in the $\Delta_1\uparrow$ band of Fe. The thin-film peaks in the low photon energy range cannot be explained by this picture and suggest that existence of *d*-like QWS's and/or the existence of partially confined QWS's around the Fermi level. The latter ones could play an important role in the coupling process in multilayer systems and influence indirectly as well as directly the GMR effect in multilayer structures. Furthermore this paper clearly illustrates the elegance of the MO measurement technique to investigate the finite-size effects of thin films: a relatively cheap and easy measurement technique. However the interpretation of the results is rather complicated.

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