Oscillatory Magneto-Optical Effect in a Au (001) Film Deposited on Fe: Experimental Confirmation of a Spin-Polarized Quantum Size Effect

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Magneto-optical response of the wedge-shaped Au ultrathin film grown on an Fe layer was investigated precisely. The magnetic circular dichroism in the reflection configuration oscillates with respect to the Au layer thickness showing superposition of several oscillations with different periods. The energy dependence of the oscillation periods is clearly explained by a concept of the spin-polarized quantum size effect in the Au layer by employing fully relativistic band calculation and electron-electron correlation. [S0031-9007(98)06304-2]

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Quantum size effects due to electron confinement in metallic layered systems give rise to striking new behaviors of various physical properties of these materials [1]. Quantum size effects are most directly evidenced by photoemission experiments [2]. At the interface between a paramagnetic metal and a ferromagnetic metal, the electrons experience a spin-dependent potential step, which leads to a spin-dependent reflection coefficient; this in turn leads to a spin-dependent spectral density *in the paramagnet*, as evidenced by spin-polarized photoemission [3]. The spectacular oscillatory behavior of the interlayer exchange coupling in magnetic multilayers [4] can be interpreted as due to such a spin-dependent quantum size effect [5].

It has been shown, both experimentally [6] and theoretically [7], that quantum size effects manifest themselves in the magneto-optical properties of ferromagnetic ultrathin films. In particular, this gives rise to oscillations of the polar Kerr rotation and ellipticity versus Fe thickness in an Fe ultrathin film on Au (001) [8]; further investigations revealed the existence of multiperiodic oscillations for this system [9].

It has also been shown that the Kerr rotation of the magnetic multilayer system exhibits some oscillatory behavior as a function of the thickness of the *paramagnetic* layer [10–14]. This is at first sight a more surprising effect, because paramagnetic layers are usually believed to be inactive with respect to magneto-optical effects (except for attenuation or optical interference effects, which cannot account for the observed behavior). The observed oscillation periods, however, were for some cases [10,12] the same as that of the interlayer magnetic coupling, but for the other cases [11,13,14] different from that. The apparent spread in the experimental results may be due partly to extrinsic features such as interface roughness, defects, etc. Systematic experimental results allowing a quantitative interpretation are still lacking.

A theoretical explanation of the oscillations of Kerr effect versus paramagnetic overlayer thickness was proposed by Bruno, Suzuki, and Chappert (hereafter referred to as BSC) [15]. The mechanism proposed by BSC may be summarized as follows. In a paramagnetic film deposited on a ferromagnetic substrate the electronic states exhibit some quantum size effects because of the confinement due to reflections on the vacuum and ferromagnet boundaries. On the vacuum side, there is total (spin-independent) reflection of the electrons, for all occupied and empty states of interest in an optics experiment. On the ferromagnet boundary, electron reflection can be total or partial (it depends upon the energy and in-plane wave vector); the reflection coefficient is spin dependent because the ferromagnet acts as a spin-dependent potential step. As a result of the spin-polarized quantum size effects, the spectral density in the paramagnetic film presents some nonzero spin polarization; the latter exhibits an oscillatory behavior as a function of the thickness of the paramagnetic layer. The periods of oscillations are related to the corresponding wave vectors of the bulk paramagnet; they usually depend quite strongly on the energy and in-plane wave vector so that the net magnetization (resulting from an integration over all energies up to the Fermi level and over all in-plane wave vectors) is usually extremely small because of strong cancellation of the various contributions. However, if there are wave vectors which span an isoenergy surface taking its stationary value with respect to the in-plane wave vector, the spin polarization of the spectral density may have non-negligible value and oscillates by a period of $2\pi/q$ (q: stationary spanning k-vector). By this mechanism, the oscillatory behavior of the magnetic interlayer coupling was dominated by several stationary wave vectors which span the Fermi surface [5]. In an optics experiment, one probes the joint spectral density of occupied and empty states separated by an energy interval equal to the photon energy, $h\nu$. If the spectral density of the initial and/or final states is spin-polarized, spin-orbit coupling leads to different absorption rates and reflection coefficients R^+ and R^- for left and right circular

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polarization of the light, i.e., to a nonvanishing magneto-optical effect in the paramagnet, which oscillates with the thickness of the paramagnetic layer. This oscillation period is determined by stationary spanning vectors on the isoenergy-difference surface which is determined by Fermi's golden rule for the photoexcitation process, i.e., $h\nu = \varepsilon_f(k) - \varepsilon_i(k)$ [15,16]. Here, $\varepsilon_f(k)$ and $\varepsilon_i(k)$ are the energies of the final and initial states of photoexcitation, respectively.

The period(s) of the oscillatory magneto-optical effect for a given photon energy depend only on the bulk band structure of the paramagnet [15]. The amplitude of the oscillatory magneto-optical effect, on the other hand, is determined by the (spin-dependent) electron reflection coefficients, and thus depends on both the paramagnet and the ferromagnet. It was suggested by BSC that the best system for a quantitative test of the theory proposed in [15] would be Au/Fe (001), because of (i) an almost perfect lattice matching, and (ii) the strong spin-orbit coupling of Au. Quantitative predictions of the periods of oscillatory magneto-optical effect for the Au/Fe (001) system have been given, on the basis of a scalar-relativistic calculation of the band structure of Au, within the local density approximation (LDA). The predicted oscillation periods exhibit a striking, unmistakable, dependence upon photon energy in the energy range experimentally accessible, which is a further advantage for a quantitative test of the theory. The purpose of the present Letter is to present an experimental test of the theory proposed by BSC. The magneto-optical effect of a Au wedge on Fe (001) has been measured precisely. A multiperiodic oscillatory variation with Au thickness has been observed. The oscillation periods, for photon energies ranging between 1.5 and 5 eV, are found in excellent quantitative agreement with the predictions of [15], provided that (i) one considers the *fully relativistic* band structure of Au, and (ii) one corrects for correlation effects on the *d* bands of Au that are not correctly described within the LDA, thus providing a clear experimental confirmation of the theory proposed by BSC [15].

The sample preparation and structural characterization has been presented in detail elsewhere [6]; thus, it will be only briefly described here. The sample has been made by ultra-high-vacuum (UHV) deposition. First, a 100 nm thick Ag (001) buffer layer, a 100 nm thick Au (001) seed layer, and a bcc-Fe (001) ultrathin film of 6 monatomic layers (ML), were successively deposited at room temperature onto a MgO (001) surface previously cleaved in air. After completion of the Ag and Au deposition, the sample was annealed at 450 and 350 °C, respectively, for several minutes. Finally, the Fe film was covered by a wedge-shaped Au (001) film. The thickness of the Au wedge (of length 20 mm) runs continuously from 6 to 37 ML. The smallest thickness (6 ML) is sufficient to prevent oxidation of the Fe film after removing the sample from the UHV chamber.

The magnetic circular dichroism (MCD) of the sample has been measured in air at room temperature in reflection in the polar configuration under a perpendicular applied magnetic field of 18.8 kOe (sufficient to saturate the sample), using a Jasco J-600 Kerr spectrometer, as shown schematically in Fig. 1. By scanning the light spot along the wedge, the variation of the MCD versus Au overlayer thickness can be studied in a very precise manner. The noise level of MCD signal was 10^{-5} at $h\nu = 3.6$ eV. This corresponds to about 0.3 mdeg noise level for the Kerr ellipticity measurement.

The raw MCD curves versus Au overlayer thickness (see Fig. 2) compose a contribution due to the MCD of the Fe film; this contribution decreases monotonously with Au overlayer thickness, due to absorption of light by the Au. This background is subtracted, as described in Ref. [13], in order to extract the contribution due to the quantum size effects. The MCD versus Au overlayer thickness D (after subtraction of the monotonous background due to Fe), for a photon energy of 3.6 eV, is shown in Fig. 3. These experimental data, i.e., oscillatory part of the MCD data, which should be proportional to the oscillatory parts of the off-diagonal element of the conductivity tensor of Au cover layer, $\sigma_{xy}^{\mathrm{osc}}$, have been fitted by a sum of several oscillatory components as predicted by BSC [15],

$$\left[\frac{R^+ - R^-}{R^+ + R^-}\right]_{\mathrm{osc}} = \mathrm{Im}\left[\frac{8\pi D}{\lambda}\,\frac{\sigma_{xy}^{\mathrm{osc}}}{\sigma_{xx}}\right] = \sum_i A_i e^{-D/\delta_i} \sin\left(\frac{2\pi D}{\Lambda_i} + \phi_i\right). \quad (1)$$

In Eq. (1), σ_{xx} is the diagonal part of the conductivity tensor of the Au buffer layer. A_i is the amplitude of the ith component, Λ_i is the corresponding period, ϕ_i is the phase, and δ_i is an attenuation length. The number of oscillatory components is not given *a priori*, but is determined by the fit; it depends on the photon energy. For the case of a photon energy of 3.6 eV, as shown in Fig. 3, the best fit is obtained by considering three distinct oscillatory components.

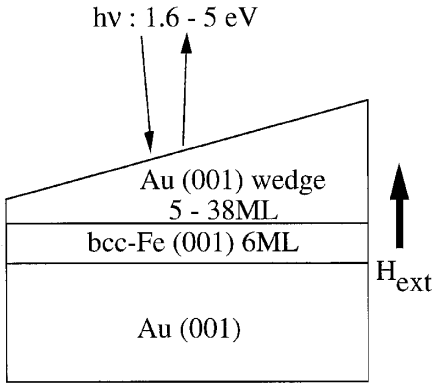

FIG. 1. Schematic view of the sample structure and configuration of the MCD measurement.

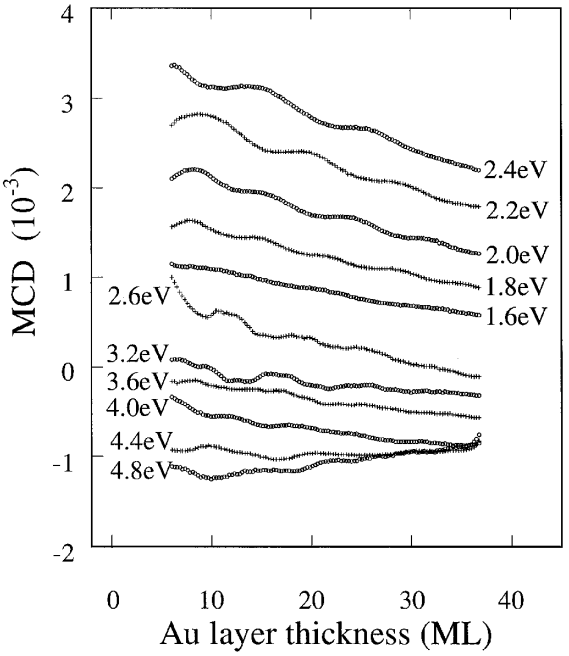

FIG. 2. MCD raw data versus Au overlayer thickness, for several photon energies. The MCD signal comprise a contribution of the Fe underlayer, absorption effect of Au coverlayer, and an additional magneto-optical activity inside Au overlayer.

The periods of oscillations of MCD for photon energies determined experimentally from the previously described procedure for photon energies ranging between 1.6 and 5 eV are shown in Fig. 4, together with the values predicted by BSC from the scalar relativistic band structure of Au calculated within the LDA (solid lines); the theoretical branch at low photon energies is due to optical transitions at the $\overline{\Gamma}$ point in 2D Brillouin zone, whereas the high photon energy branches correspond to transitions at the $\overline{M}$ point (see Ref. [15] for details) [17]. Although some similarity between experimental and theoretical results is apparent in Fig. 4, there is some strong discrepancy so that the comparison cannot be claimed to be really satisfactory.

In fact, the approximations made by BSC for the sake of simplicity when calculating explicitly the oscillation periods for Au/Fe (001) [15] must be released if one wants to obtain quantitative results to be compared with experiments. These approximations are (i) the LDA, and (ii) the scalar relativistic approximation (i.e., the neglect of the spin-orbit coupling in the band structure calculations). We have released the latter approximation by performing *fully relativistic* band structure calculations; this leads to spin-orbit splittings in the band structure (which for Au are quite large and cannot be neglected). As a result, all the branches shown by solid lines in Fig. 4 are split into three different branches, with a maximum splitting of the order of 1 eV.

The errors due to the LDA, on the other hand, are a much more serious problem, and, in fact, cannot be corrected rigorously. Thus, one has to rely on more qualitative arguments. For noble metals, it is well known that the LDA gives very good results for the empty *sp* band, but rather poor ones for the closed-shell *d* bands

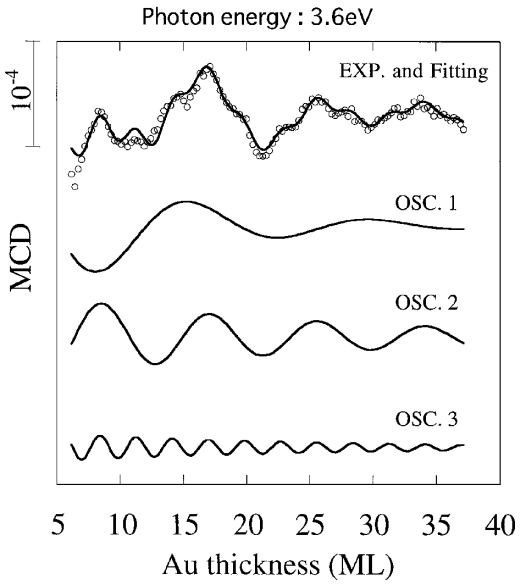

FIG. 3. MCD versus Au overlayer thickness, for a photon energy of 3.6 eV. The open points are the experimental data, after subtraction of a smooth background curve due to the MCD of the underlying Fe film. The solid line is a fit (see text for details) comprising three oscillatory components shown separately below.

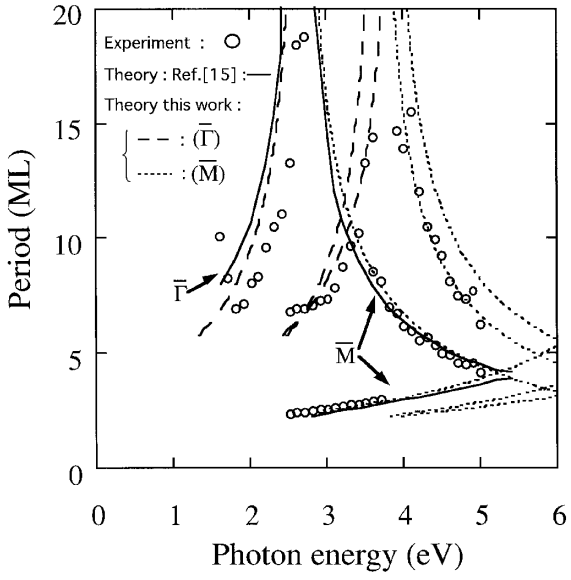

FIG. 4. Periods of MCD oscillations versus photon energy. Open circles: Experimental results. Solid lines: Theoretically predicted periods, as obtained from the scalar relativistic band structure of Au calculated within the LDA [15]. Broken and dotted lines: Theoretically predicted periods, as obtained from the fully relativistic LDA band structure of Au with correction for correlation effects on the binding energies of *d* bands (see text for details).

[18]; in particular, the LDA underestimates the binding energy of the valence d bands of noble metals by about 1 eV, as compared with photoemission results [19]. This is due to correlation effects in the closed d shell, which the LDA fails to describe correctly. In our case, where we are interested in optical transitions in Au between occupied d states and empty p states, this leads to underestimating optical transition energies by about 1 eV. Thus, we argue that (at least in a first approximation) we can correct for these correlation effects by a rigid upward shift of 1 eV of the transition energies obtained from the LDA calculations.

The theoretical predictions obtained by the previously described procedure (i.e., fully relativistic band structure calculations, plus rigid shift of $d \rightarrow p$ optical transition energies) are compared to the experimental results in Fig. 4 also (broken and dotted lines). The agreement appears excellent: Except for a few points at low photon energies, all the experimental points are found to lie on the predicted theoretical curves. The fact that the measured periods follow the dispersion calculated from the band structure of the Au clearly excludes the possibility of explaining the observed oscillations of magneto-optical effects by optical transitions taking place in the Fe itself. Thus, these results provide an unambiguous experimental proof of the validity of the mechanism proposed by BSC [15], which attributes the oscillations of magneto-optical properties versus paramagnetic overlayer thickness to a spin-polarized quantum size effect *in the paramagnetic overlayer*.

In order to deepen further our understanding of magneto-optical effects in ultrathin layered systems, it would be desirable to compute directly the magneto-optical properties of the model system Au/Fe (001) from first principles, and to investigate their oscillatory behavior. Performing such calculations is a rather big challenge, but in view of the remarkable progress made in this field recently [20], this seems feasible.

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