

# Magneto-optical Kerr effect in a paramagnetic overlayer on a ferromagnetic substrate: A spin-polarized quantum size effect

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The magneto-optical properties of a paramagnetic overlayer on a ferromagnetic substrate are discussed. The spin-dependent electron confinement in the overlayer due to the ferromagnetic substrate gives rise to spin-polarized quantum size effects in the paramagnetic overlayer. It is shown that the latter induce a nonvanishing Kerr effect in the overlayer, which oscillates with overlayer thickness. The mechanism of Kerr effect in this case is rather different from the one of Kerr effect in a bulk ferromagnet. The periods of oscillations are related to the bulk band structure of the overlayer material. The theory is applied to the case of Au/Fe(001); the calculated period of Kerr effect oscillations versus Au thickness is found to vary in a dramatic manner with photon energy in the visible-UV range.

## I. INTRODUCTION

Quantum size effects manifest themselves in solids having one (at least) dimension comparable with the electron wavelength. They are mostly investigated in thin films, where electrons are confined along the normal direction only. In metallic films, the characteristic length is the Fermi wavelength, of the order of the interatomic distance, so that quantum size effects show up only for extremely thin films.

The investigation of quantum size effects in magnetic ultrathin films and multilayers is currently attracting considerable attention.<sup>1-3</sup> The spectacular phenomenon of oscillatory interlayer exchange coupling,<sup>4</sup> in particular, may be interpreted as a quantum size effect.<sup>5-7</sup>

Recently, it was shown that quantum size effects manifest also themselves in the magneto-optical properties of ferromagnetic ultrathin films.<sup>8,9</sup> This gives rise to oscillations of the polar Kerr rotation and ellipticity versus Fe thickness in an Fe ultrathin film on Au(001).<sup>10</sup> Furthermore, multiperiodic oscillations have been found in the latter system.<sup>11</sup> A quantitative theory of this effect in terms of quantum confinement of electrons in the Fe layer has been proposed recently; the theoretical oscillation periods are in good agreement with the experimental ones.<sup>11</sup>

In this paper, we present a theoretical investigation of the Kerr effect in a *paramagnetic* overlayer on a ferromagnetic substrate. This is motivated by the recent discovery of oscillations of the Kerr rotation versus Au overlayer thickness in the system Au(111)/Co(0001)/Au(111).<sup>12</sup> As suggested in Ref. 12, we shall show that this behavior may be attributed to the Kerr effect in the *paramagnetic* Au overlayer. However, in contrast to the case of a ferromagnet, where the Kerr effect is due to simultaneous occurrence of exchange splitting and spin-orbit coupling, the Kerr effect in the paramagnetic overlayer (which has zero exchange splitting) is due to a rather different mechanism: It is actually a quantum size effect due to spin-dependent electron reflectivity at the paramagnet-ferromagnet interface.

In Sec. II, general considerations on magneto-optical effects are given. Then, in Sec. III, we discuss the quantum

size effects due to electron confinement in an overlayer. The mechanism of the Kerr effect in a paramagnetic overlayer on a ferromagnetic substrate is presented in Sec. IV. Finally, Sec. V is devoted to the discussion of a realistic system, namely, Au/Fe(001).

## II. KERR EFFECT AND OPTICAL CONDUCTIVITY TENSOR

It is well known that the magneto-optical Kerr effect is related to off-diagonal components of the optical conductivity tensor. In this paper, we will restrict ourselves to the case of the polar Kerr effect, with the magnetization and light incidence direction perpendicular to the film plane. Furthermore, we shall consider only systems which have at least threefold rotational in-plane symmetry. The conductivity tensor then is of the form

$$\sigma(\omega) = \begin{pmatrix} \sigma_{xx}(\omega) & \sigma_{xy}(\omega) & 0 \\ -\sigma_{xy}(\omega) & \sigma_{xx}(\omega) & 0 \\ 0 & 0 & \sigma_{zz}(\omega) \end{pmatrix}, \quad (1)$$

where the  $z$  axis is taken along the normal direction. The Kerr rotation angle  $\theta_K$  and the Kerr ellipticity  $\varepsilon_K$  may be combined to form the complex Kerr angle<sup>13</sup>

$$\phi_K \equiv \theta_K + i\varepsilon_K. \quad (2)$$

For a film of thickness  $D$  on a substrate, the Kerr rotation angle is<sup>8</sup>

$$\phi_K = \frac{i\sigma_{xy}}{\sigma_{xx}^s} \frac{4\pi D}{\lambda}, \quad (3)$$

where  $\sigma_{xx}^s$  is the optical conductivity of the substrate and  $\lambda = 2\pi c/\omega$  the wavelength of the light in vacuum; this expression is valid when  $D \ll \lambda$ .

The real part  $\sigma'(\omega)$  and imaginary part  $\sigma''(\omega)$  of the conductivity tensor are, respectively, even and odd with respect to  $\omega$  and are linked by the well-known Kramers-Krönig relations.<sup>14</sup> The expression of the conductivity tensor in terms of the microscopic electronic structure may be

obtained from the Fermi golden rule<sup>15,16</sup> or by using the Kubo formalism.<sup>17</sup> The dissipative part of the off-diagonal component of the conductivity tensor (for  $\omega > 0$ ) is then found to be<sup>15</sup>

$$\sigma''_{xy}(\omega) = \frac{\pi e^2}{4\hbar\omega m^2\Omega} \sum_{i,f} f(\varepsilon_i)[1-f(\varepsilon_f)] \times [|\langle i|p_-|f\rangle|^2 - |\langle i|p_+|f\rangle|^2] \delta(\omega_{fi} - \omega), \quad (4)$$

where  $p_{\pm} \equiv p_x \pm ip_y$ ,  $f(\varepsilon)$  is the Fermi-Dirac function,  $\Omega$  the total volume, and  $\hbar\omega_{fi} \equiv \varepsilon_f - \varepsilon_i$ .

The above expression is interpreted straightforwardly in terms of the absorption of a photon by an electron transiting between an occupied initial state  $|i\rangle$  and an unoccupied final state  $|f\rangle$ ; the factor  $\delta(\omega_{fi} - \omega)$  expresses the condition of energy conservation. The matrix elements  $\langle i|p_-|f\rangle$  and  $\langle i|p_+|f\rangle$  correspond to dipolar electric transitions, for right and left circularly polarized light, respectively. Clearly,  $\sigma''_{xy}(\omega)$  is proportional to the difference of absorption probability for right and left circularly polarized light. Similarly,  $\sigma''_{xx}(\omega)$  is proportional to the average absorption. The corresponding dispersive components,  $\sigma'_{xx}(\omega)$  and  $\sigma'_{xy}(\omega)$  are obtained by using the Kramers-Krönig relations.<sup>14</sup>

Before considering the case of overlayers, it is instructive to discuss the mechanism of the Kerr effect in bulk materials. In a three-dimensional system, the matrix elements vanish unless the initial state  $|i\rangle$  and final state  $|f\rangle$  of the optical transition have the same wave vector  $\mathbf{k}$  (as done usually, we neglect the much smaller wave vector  $\mathbf{K}$  of the photon); i.e., only vertical optical transitions are allowed. This is expressed by

$$|\langle \mathbf{k}, i|p_{\pm}|\mathbf{k}', f\rangle|^2 = \frac{8\pi^3}{\Omega_0} \delta^{(3)}(\mathbf{k} - \mathbf{k}') |\langle \mathbf{k}, i|p_{\pm}|\mathbf{k}, f\rangle|^2, \quad (5)$$

where  $\Omega_0$  is the atomic volume.

In addition, the selection rules for electric dipolar transitions must be satisfied, i.e.,

$$\Delta l = \pm 1, \quad (6a)$$

$$\Delta m_l = \pm 1. \quad (6b)$$

The first selection rule implies that only transitions between  $s$  and  $p$  levels or between  $p$  and  $d$  levels (for transition metals) are allowed. For the second selection rule, the transitions with  $\Delta m_l = +1$  and  $\Delta m_l = -1$  correspond to left and right circularly polarized light, respectively.

As an example, let us consider a transition between a doubly degenerate  $d_{xz,yz}$  level ( $l=2, m_l = \pm 1$ ) and a  $p_z$  level ( $l=1, m_l=0$ ). The majority and minority spin  $d$  levels, in the ferromagnet, are separated by the exchange splitting  $\Delta_{ex}$ . Due to spin-orbit coupling, the orbital degeneracy of the  $d_{xz,yz}$  levels is lifted, and the latter are split into  $d_{(x+iy)z}$  (having  $m_l = +1$ ) and  $d_{(x-iy)z}$  (having  $m_l = -1$ ) levels. For spin up (i.e., majority spin), the level with  $m_l = +1$  has a higher energy, whereas for spin down (i.e., minority spin), the converse holds. This is sketched in Fig. 1. From this picture, it appears clearly that, in a bulk ferromagnet, the Kerr effect arises from the simultaneous occurrence of exchange splitting and spin-orbit coupling.

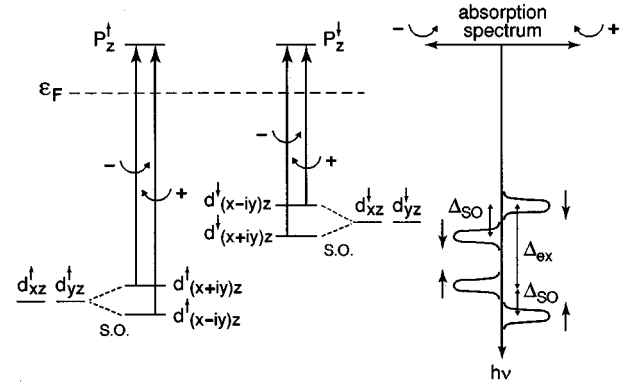


FIG. 1. Sketch of the energy levels in a bulk ferromagnet, showing the electric dipolar optical transitions for left and right circular light. The corresponding absorption spectra versus photon energy  $h\nu$  are shown on the right; the solid and dashed lines correspond to spin up and spin down transitions, respectively.

By comparison, the case of a bulk paramagnet is sketched in Fig. 2, showing that the cancellation of majority and minority spin contributions leads to a vanishing Kerr effect. More generally, in a bulk paramagnet, the absence of the Kerr effect is due to the fact that

$$|\langle i\uparrow|p_+|f\uparrow\rangle| = |\langle i\downarrow|p_-|f\downarrow\rangle|, \quad (7a)$$

$$|\langle i\uparrow|p_-|f\uparrow\rangle| = |\langle i\downarrow|p_+|f\downarrow\rangle|. \quad (7b)$$

### III. QUANTUM SIZE EFFECT IN AN OVERLAYER

Let us now consider the effect of electron confinement in a paramagnetic overlayer on a ferromagnetic substrate. The discussion follows that given in earlier publications.<sup>6,18</sup>

The system under consideration consists of a paramagnetic layer of thickness  $D$  bounded on one side by a ferromagnetic material and by vacuum on the other side. The electron states of the bulk paramagnet are noted,  $|\mathbf{k}_{\parallel}, k_{\perp}^{\pm}, n, \sigma\rangle$ , where  $\mathbf{k}_{\parallel}$  is the in-plane wave vector,  $k_{\perp}^{\pm}$  the perpendicular wave vector,  $n$  the band index, and  $\sigma$  the spin index (the spin quantization axis is chosen parallel to the magnetization of the ferromagnetic substrate, i.e., along the  $z$  axis). These states have an energy  $\varepsilon_n(\mathbf{k}_{\parallel}, k_{\perp}^{\pm})$ , independent of the spin. The upper indices  $+$  and  $-$  correspond to states

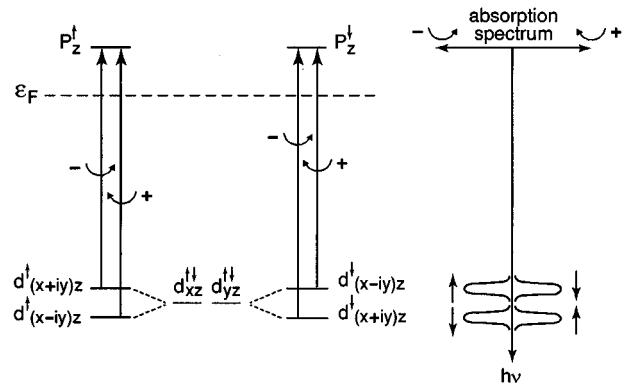


FIG. 2. Same as Fig. 1, for a bulk paramagnet.

having a positive and negative velocity  $v_{\perp}$  along the  $z$  axis, respectively.

The effect of confinement can be described in terms of the electron states of the bulk paramagnet and of the reflection coefficients  $r^{\uparrow}$  and  $r^{\downarrow}$  at the paramagnet-ferromagnet interface, respectively for majority and minority spin, and  $r^{\text{vac}}$  at the paramagnet-vacuum interface. The reflection coefficients, of course, depend on the energy and in-plane wave vector.

Let us consider an electron traveling in the overlayer. Due to reflections on the boundaries, interferences take place and lead to changes in the density of states: Constructive (destructive) interferences lead to an increase (a decrease) of the density of states. As was shown in Ref. 18, the change  $\Delta N_{\sigma}(\varepsilon, \mathbf{k}_{\parallel})$  of the integrated density of states for a state of energy  $\varepsilon$ , in-plane wave vector  $\mathbf{k}_{\parallel}$ , and spin  $\sigma$ , due to confinement, is given by

$$\Delta N_{\sigma}(\varepsilon, \mathbf{k}_{\parallel}) = -\frac{1}{\pi} \text{Im} \ln [1 - r^{\text{vac}} r^{\sigma} e^{i(k_{\perp}^{+} - k_{\perp}^{-})D}]. \quad (8)$$

The corresponding density of states may be written as

$$n_{\sigma}(\varepsilon, \mathbf{k}_{\parallel}) = n_0(\varepsilon, \mathbf{k}_{\parallel}) g_{\sigma}(\varepsilon, \mathbf{k}_{\parallel}), \quad (9)$$

where  $n_0(\varepsilon, \mathbf{k}_{\parallel})$  is the density of states of the bulk material and  $g_{\sigma}(\varepsilon, \mathbf{k}_{\parallel})$  expresses the relative change of spectral weight due to confinement. If we neglect the energy dependence of the product  $r^{\text{vac}} r^{\sigma}$  as compared to the one of the exponential factor (this is valid when  $D$  is large enough), we obtain

$$\begin{aligned} g_{\sigma}(\varepsilon, \mathbf{k}_{\parallel}) &= \text{Re} \left[ \frac{1 + r^{\text{vac}} r^{\sigma} e^{i(k_{\perp}^{+} - k_{\perp}^{-})D}}{1 - r^{\text{vac}} r^{\sigma} e^{i(k_{\perp}^{+} - k_{\perp}^{-})D}} \right] \\ &= \text{Re} \left[ 1 + \frac{2r^{\text{vac}} r^{\sigma} e^{i(k_{\perp}^{+} - k_{\perp}^{-})D}}{1 - r^{\text{vac}} r^{\sigma} e^{i(k_{\perp}^{+} - k_{\perp}^{-})D}} \right]. \end{aligned} \quad (10)$$

The effect of confinement is to modulate periodically the spectral weight of a given state. As the reflection coefficient at the paramagnet-ferromagnet interface depends on the spin of the incident electron, the modulation of the spectral weight is spin dependent. The confinement strength is measured by  $|r^{\text{vac}} r^{\uparrow(\downarrow)}|$ . The variation of the weighting factor  $g$  versus layer thickness is shown in Fig. 3, for various values of the confinement strength. In the case of a weak confinement, one obtains a sinelike modulation of the spectral weight. In the opposite limit of complete confinement ( $|r^{\text{vac}} r^{\uparrow(\downarrow)}| = 1$ ), one has quantization of the allowed states, and the weighting factor  $g$  is zero, except for quantized states, where it behaves like a  $\delta$  function.

Obviously, one has  $|r^{\text{vac}}| = 1$ , so that we can write

$$r^{\text{vac}} = e^{i\phi^{\text{vac}}}, \quad (11)$$

where  $\phi^{\text{vac}}$  is the phase shift for reflection on the paramagnet-vacuum interface. On the other hand, the reflection coefficient at the paramagnet-ferromagnet interface,  $r^{\uparrow(\downarrow)}$ , has a modulus equal to 1 or smaller than 1, depending on the existence or nonexistence of a local gap in the ferromagnet band structure.

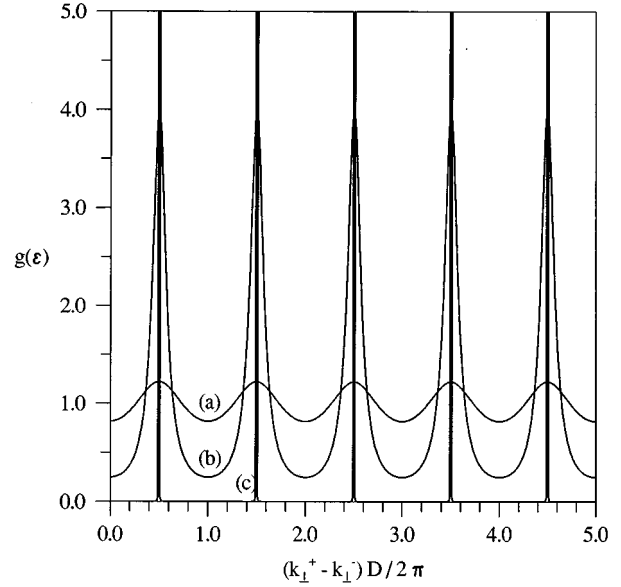


FIG. 3. Variation of the relative change of spectral weight,  $g(\varepsilon)$ , due to confinement, as a function of the layer thickness  $D$ . The curves labeled (a), (b), and (c) correspond, respectively, to the cases of weak confinement ( $|r^{\text{vac}} r^{\uparrow(\downarrow)}| = 0.1$ ), intermediate confinement ( $|r^{\text{vac}} r^{\uparrow(\downarrow)}| = 0.6$ ), and total confinement ( $|r^{\text{vac}} r^{\uparrow(\downarrow)}| = 1$ ).

#### IV. MECHANISM OF THE KERR EFFECT

The change of spectral weight due to confinement induces a change in the matrix elements of  $p_{\pm}$ . Clearly, this change must be of the form

$$|\langle i | p_{\pm} | f \rangle|^2 \approx |\langle i | p_{\pm} | f \rangle_0|^2 g(i) g(f), \quad (12)$$

where  $|\langle i | p_{\pm} | f \rangle_0|^2$  is the matrix element for the bulk material and where  $g(i)$  and  $g(f)$  are the relative changes of spectral weight due to confinement for the initial and final states, respectively.

Furthermore, because we are considering a slab of finite thickness  $D$ , the strict conservation of  $k_{\perp}$  for an optical transition is relaxed, and nonvertical transitions with a change  $\Delta k_{\perp}$  of  $k_{\perp}$  of the order of  $\pi/D$  are allowed.<sup>19</sup> Thus, in Eq. (5), we have to perform the substitution

$$\delta(k_{i\perp} - k_{f\perp}) \rightarrow F(k_{i\perp} - k_{f\perp}), \quad (13)$$

where  $F(k_{i\perp} - k_{f\perp})$  is significantly different from zero only for  $|k_{i\perp} - k_{f\perp}| < \pi/D$ . A reasonable choice is a Lorentzian, i.e.,

$$F(k_{i\perp} - k_{f\perp}) = \frac{D}{\pi^2 + (k_{i\perp} - k_{f\perp})^2 D^2}. \quad (14)$$

Thus, the matrix element of  $p_{\pm}$  between an initial state  $|\mathbf{k}_{\parallel}, k_{i\perp}, n_i, \sigma\rangle$  and a final state  $|\mathbf{k}_{\parallel}, k_{f\perp}, n_f, \sigma\rangle$ , with  $k_{i\perp} = k_{\perp} + \Delta k_{\perp}/2$  and  $k_{f\perp} = k_{\perp} - \Delta k_{\perp}/2$ , becomes

$$|\langle \mathbf{k}_{\parallel}, k_{i\perp}, n_i, \sigma | p_{\pm} | \mathbf{k}_{\parallel}, k_{f\perp}, n_f, \sigma \rangle|^2 \approx |\langle \mathbf{k}_{\parallel}, k_{\perp}, n_i, \sigma | p_{\pm} | \mathbf{k}_{\parallel}, k_{\perp}, n_f, \sigma \rangle_0|^2 \frac{2\pi}{d} F(\Delta k_{\perp}) g_{\sigma}(\mathbf{k}_{\parallel}, k_{i\perp}, n_i) g_{\sigma}(\mathbf{k}_{\parallel}, k_{f\perp}, n_f). \quad (15)$$

Since the reflection coefficients for majority and minority spins at the paramagnet-ferromagnet interface,  $r_{\uparrow}$  and  $r_{\downarrow}$ , are different, the change of spectral weight in the paramagnet due to confinement is spin dependent. Thus, relations (7a) and (7b) no longer hold, so that the absorption spectra for left and right circularly polarized light are no longer the same: i.e., one has a nonvanishing Kerr effect in the paramagnetic overlayer.

Let us emphasize that the mechanism by which the Kerr effect takes place in a paramagnetic overlayer on a ferromagnetic substrate is rather different from the one at work in a bulk ferromagnet: As noted in Sec. II, the Kerr effect takes place in a bulk ferromagnet because the absorption lines for left and right circularly polarized light occur at different photon energies, due to the exchange splitting. In contrast to this, in a paramagnetic overlayer on a ferromagnetic substrate, the absorption lines for left and right circularly polarized light occur for the same photon energy, but have different intensities, due to the spin-dependent quantum size effect in the paramagnetic overlayer.

The expression of  $\sigma''_{xy}(\omega)$  becomes

$$\sigma''_{xy}(\omega) = \frac{\pi e^2}{4\hbar \omega m^2} \sum_{n_i, n_f} \frac{1}{8\pi^3} \int d^2 \mathbf{k}_{\parallel} \int_{-\pi/d}^{\pi/d} dk_{i\perp} \int_{-\pi/d}^{\pi/d} dk_{f\perp} f(\varepsilon_i) [1 - f(\varepsilon_f)] [|p_{-if}^{\uparrow 0}|^2 - |p_{+if}^{\uparrow 0}|^2] F(k_{f\perp} - k_{i\perp}) \times [g_{\uparrow}(i)g_{\uparrow}(f) - g_{\downarrow}(i)g_{\downarrow}(f)] \frac{1}{\pi} \frac{1/\tau}{(\omega_{fi} - \omega)^2 + 1/\tau^2}, \quad (16)$$

where we have used the shorthand notation

$$|p_{-if}^{\uparrow 0}|^2 - |p_{+if}^{\uparrow 0}|^2 \equiv \frac{\Omega}{\Omega_0} [|\langle \mathbf{k}_{\parallel}, k_{\perp}, n_i, \uparrow | p_{-} | \mathbf{k}_{\parallel}, k_{\perp}, n_f, \uparrow \rangle_0|^2 - |\langle \mathbf{k}_{\parallel}, k_{\perp}, n_i, \uparrow | p_{+} | \mathbf{k}_{\parallel}, k_{\perp}, n_f, \uparrow \rangle_0|^2] \quad (17)$$

and

$$g_{\uparrow(\downarrow)}(i) \equiv g_{\uparrow(\downarrow)}(\mathbf{k}_{\parallel}, k_{i\perp}, n_i), \quad (18a)$$

$$g_{\uparrow(\downarrow)}(f) \equiv g_{\uparrow(\downarrow)}(\mathbf{k}_{\parallel}, k_{f\perp}, n_f), \quad (18b)$$

and where we have performed the substitution

$$\delta(\omega_{fi} - \omega) \rightarrow \frac{1}{\pi} \frac{1/\tau}{(\omega_{fi} - \omega)^2 + 1/\tau^2}, \quad (19)$$

to take into account the finite lifetime  $\tau$  of the excitation.

Let us now discuss the variation of  $\sigma''_{xy}(\omega)$  versus the overlayer thickness  $D$ . For simplicity, we shall consider the case of weak confinement, i.e.,  $|r^{\text{vac}} r^{\uparrow(\downarrow)}| \ll 1$ , and large overlayer thickness  $D$ . For weak confinement, one has

$$g_{\sigma}(\varepsilon, \mathbf{k}_{\parallel}) \approx \text{Re} [1 + 2r^{\text{vac}} r^{\uparrow(\downarrow)} e^{i(k_{\perp}^{+} - k_{\perp}^{-})D}], \quad (20)$$

so that

$$g_{\uparrow}(i)g_{\uparrow}(f) - g_{\downarrow}(i)g_{\downarrow}(f) \approx 4 \text{Re}(r_i^{\text{vac}} \Delta r_i e^{iq_{i\perp} D} + r_f^{\text{vac}} \Delta r_f e^{iq_{f\perp} D}), \quad (21)$$

where

$$\Delta r \equiv \frac{r^{\uparrow} - r^{\downarrow}}{2} \equiv |\Delta r| e^{i\psi} \quad (22)$$

is the spin asymmetry of reflection coefficient at the paramagnet-ferromagnet interface and where

$$q_{\perp} \equiv k_{\perp}^{+} - k_{\perp}^{-}. \quad (23)$$

The indices  $i$  and  $f$  indicate that the quantity under consideration is calculated for the initial and final states of the optical transition, respectively.

We see that  $\sigma''_{xy}(\omega)$  presents an oscillatory behavior with respect to  $D$ . Clearly, we can identify oscillatory terms due to confinement of the initial and final states, respectively, which can be treated separately.

In the limit of large thickness  $D$ , the integrals over  $k_{i\perp}$  and  $k_{f\perp}$  are easily performed by complex-plane integration methods, and one obtains

$$\sigma''_{xy} = \frac{\pi e^2}{4\hbar \omega m^2} \sum_{n_i, n_f} \frac{1}{8\pi^3} \int d^2 \mathbf{k}_{\parallel} f(\varepsilon_i) [1 - f(\varepsilon_f)] [|p_{-if}^{\uparrow 0}|^2 - |p_{+if}^{\uparrow 0}|^2] \frac{1}{|v_{i\perp} - v_{f\perp}|} \times (-4) \text{Re}[r_i^{\text{vac}} \Delta r_i e^{iq_{i\perp} D} \beta_{if} e^{-D/D_{if}} + r_f^{\text{vac}} \Delta r_f e^{iq_{f\perp} D} \beta_{fi} e^{-D/D_{fi}}], \quad (24)$$

where

$$\beta_{if} \equiv \exp\left(\frac{-\pi|\alpha_i v_{f\perp}|}{|v_{i\perp} - v_{f\perp}|}\right) \quad (25)$$

and

$$D_{if} \equiv \frac{|v_{i\perp} - v_{f\perp}|\tau}{|\alpha_i|}, \quad (26)$$

with the dimensionless quantity  $\alpha_i$  defined by<sup>20</sup>

$$\alpha_i \equiv \frac{dq_{i\perp}}{dk_{i\perp}}. \quad (27)$$

The definitions of  $\beta_{fi}$ ,  $D_{fi}$ , and  $\alpha_f$  are obtained from those of  $\beta_{if}$ ,  $D_{if}$ , and  $\alpha_i$ , respectively, by interchanging the indices  $i$  and  $f$ . In Eq. (24), the indices  $i$  and  $f$  refer to the states  $|\mathbf{k}_{\parallel}k_{\perp}n_i\rangle$  and  $|\mathbf{k}_{\parallel}k_{\perp}n_f\rangle$  satisfying  $\omega_{fi} = \omega$ ; if such a pair of states does not exist for a given value of  $\mathbf{k}_{\parallel}$ , the integrand vanishes.

The interpretation of Eq. (24) is rather simple. The first and second terms correspond, respectively, to confinement in the initial state  $i$  and in the final state  $f$ . Since both terms are similar, we shall focus on the former and explain the physical meaning of the various factors. The factor  $|v_{i\perp} - v_{f\perp}|^{-1}$  is essentially the joint density of states, giving the weight of the transition  $i \rightarrow f$  in the absorption spectrum. The factor  $r_i^{\text{vac}} \Delta r_i e^{iq_{i\perp}D}$  expresses the spin asymmetry of the confinement in the initial state  $i$ . The possibility of having nonver-

tical transitions with a change  $\Delta k_{\perp}$  of the order of  $\pi/D$  leads to a blurring of the oscillations, which is responsible for the reduction factor  $\beta_{if}$ . Finally, the factor  $e^{-D/D_{if}}$  results from the blurring due to the finite lifetime  $\tau$ .

Then we have to integrate over  $\mathbf{k}_{\parallel}$ . Since the contributions of the various  $\mathbf{k}_{\parallel}$  oscillate with different periods, they cancel each other except near vectors  $\mathbf{k}_{\parallel}^{\mu}$  where  $q_{i\perp}$  is stationary with respect to  $\mathbf{k}_{\parallel}$ . This is completely analogous to the problem of oscillatory interlayer exchange coupling.<sup>18,21</sup> In the limit of large overlayer thickness, the integration over  $\mathbf{k}_{\parallel}$  is easily performed by using the stationary phase approximation. Let us label with an index  $\mu$  the vectors  $\mathbf{k}_{\parallel}$  corresponding to stationary  $q_{i\perp}$  and with an index  $\nu$  those corresponding to stationary  $q_{f\perp}$ . Near  $\mathbf{k}_{\parallel}^{\mu}$ ,  $q_{i\perp}$  may be expanded as

$$q_{i\perp} \approx q_{i\perp}^{\mu} - \frac{(k_{ix} - k_{ix}^{\mu})^2}{\kappa_{ix}^{\mu}} - \frac{(k_{iy} - k_{iy}^{\mu})^2}{\kappa_{iy}^{\mu}}, \quad (28)$$

where the crossed terms have been eliminated by a suitable choice of the axes  $x$  and  $y$ . One also introduces

$$\kappa_i^{\mu} \equiv |\kappa_{ix}^{\mu} \kappa_{iy}^{\mu}|^{1/2} \quad (29)$$

and  $\delta_i^{\mu}$ , respectively, equal to 0,  $\pi/2$ , or  $\pi$ , when  $q_{i\perp}$  is a maximum, a saddle point, or a minimum, at  $\mathbf{k}_{\parallel}^{\mu}$ . The development of  $q_{f\perp}$  near  $\mathbf{k}_{\parallel}^{\nu}$  is analogous.

The final result is

$$\begin{aligned} \sigma_{xy}^{\mu}(\omega) = & \frac{-e^2}{8\pi\hbar\omega m^2 D} \sum_{\mu} f(\varepsilon_i^{\mu}) [1 - f(\varepsilon_f^{\mu})] [|p_{-if}^{\uparrow 0}|^2 - |p_{+if}^{\uparrow 0}|^2]_{\mu} \frac{|\Delta r_i^{\mu}| \kappa_i^{\mu} \beta_{if}^{\mu} e^{-D/D_{if}^{\mu}}}{|v_{i\perp}^{\mu} - v_{f\perp}^{\mu}|} \sin(q_{i\perp}^{\mu} D + \phi_i^{\mu} + \psi_i^{\mu} + \delta_i^{\mu}) \\ & + \frac{-e^2}{8\pi\hbar\omega m^2 D} \sum_{\nu} f(\varepsilon_i^{\nu}) [1 - f(\varepsilon_f^{\nu})] [|p_{-if}^{\uparrow 0}|^2 - |p_{+if}^{\uparrow 0}|^2]_{\nu} \frac{|\Delta r_f^{\nu}| \kappa_f^{\nu} \beta_{if}^{\nu} e^{-D/D_{if}^{\nu}}}{|v_{i\perp}^{\nu} - v_{f\perp}^{\nu}|} \sin(q_{f\perp}^{\nu} D + \phi_f^{\nu} + \psi_f^{\nu} + \delta_f^{\nu}). \quad (30) \end{aligned}$$

The periods of oscillations are determined by the vectors  $q_{i\perp}^{\mu}$  and  $q_{f\perp}^{\nu}$  satisfying the stationarity criterium given above. The phase shifts  $\phi_i^{\mu}$  and  $\phi_f^{\nu}$  are due to the reflection on the vacuum, while  $\psi_i^{\mu}$  and  $\psi_f^{\nu}$  arise from the reflection on the ferromagnet.

The strength of the oscillatory components of the Kerr effect in the paramagnetic overlayer depends on several factors. The left-right asymmetry factor

$$[|p_{-if}^{\uparrow 0}|^2 - |p_{+if}^{\uparrow 0}|^2] \quad (31)$$

is proportional to the spin-orbit coupling, so that large effects are expected for paramagnetic materials such as gold, having a high atomic number and, hence, a large spin-orbit coupling. Next, in order to obtain a strong quantum size effect, a large spin asymmetry  $\Delta r$  of the reflection coefficient at the paramagnet-ferromagnet interface is required, for the initial and/or final state; this is achieved, for example, for states of the paramagnet corresponding to a local gap for one of the exchange-split bands of same symmetry in the ferromagnet.

Next, for the case of confinement in the initial (respectively, final) state, in order to maximize the factors  $\beta_{if}^{\mu}$  and  $e^{-D/D_{if}^{\mu}}$  (or  $\beta_{if}^{\nu}$  and  $e^{-D/D_{if}^{\nu}}$  for the case of confinement in the final state), the confined state must be one of large velocity  $v_{\perp}$ , whereas the other must have a small velocity.

## V. DISCUSSION OF A REALISTIC CASE: Au/Fe(001)

As discussed above, gold with its large spin-orbit coupling is a good candidate for observing magneto-optical effects in overlayers. The case of a Au(111) overlayer on Co(0001) has been investigated experimentally, and oscillations of the Kerr effect versus Au overlayer thickness have been observed.<sup>12</sup> However, this system is difficult to treat theoretically; this is due in particular to the large lattice mismatch between Au and Co, which causes many dislocations being present at the interface. Furthermore, the fcc lattice is not symmetric with respect to (111) planes, which makes the theoretical analysis, and hence the comparison with experimental data, more problematic.

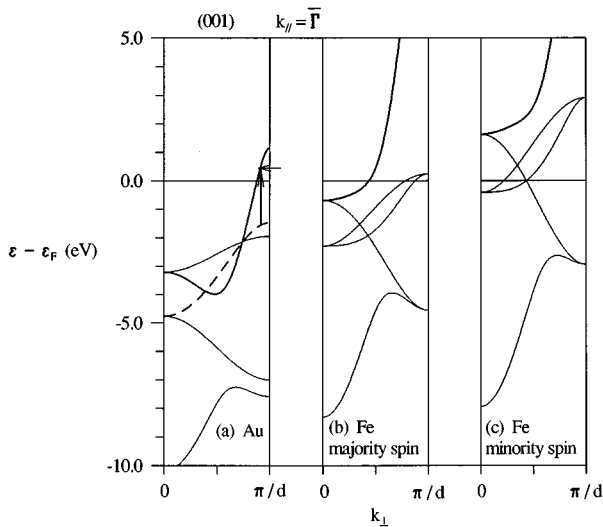


FIG. 4. Band dispersion vs  $k_{\perp}$  of Au (a) and majority (b) and minority (c) spin Fe, for the center  $\bar{\Gamma}$  of the (001) two-dimensional Brillouin zone. Explanations are given in the text.

Below, we shall apply the above theory to the case of a Au(001) overlayer on a Fe(001) substrate. There is an almost perfect lattice matching between Au(001) and bcc Fe(001); furthermore, Fe whiskers can be used as Fe(001) substrates of unrivalled crystalline quality. These features allowed one to observe the multiperiodic oscillatory interlayer exchange coupling in Fe/Au/Fe up to Au spacer thicknesses as large as 60 atomic layers (AL),<sup>22</sup> with periods in perfect agreement with the theoretical predictions.<sup>21</sup> Moreover, inverse photoemission investigations have given direct evidence of quantum size effects in Au(001) overlayers on Fe(001).<sup>23</sup> All these considerations make the Au/Fe(001) system one of choice for a quantitative experimental test of the present theory.

As was discussed in Ref. 18 for the analogous problem of oscillatory interlayer exchange coupling, the stationary vectors  $q_{i\perp}^{\mu}$  or  $q_{f\perp}^{\nu}$  are found primarily at high-symmetry points of the two-dimensional Brillouin zone because of symmetry

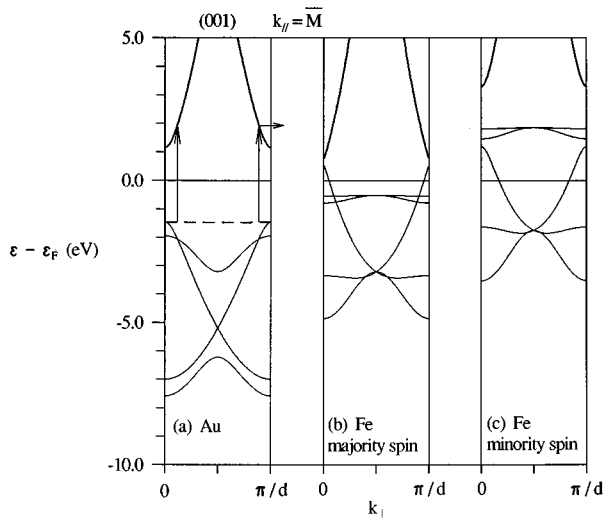


FIG. 5. Same as Fig. 4, for the point  $\bar{M}$  of the (001) two-dimensional Brillouin zone.

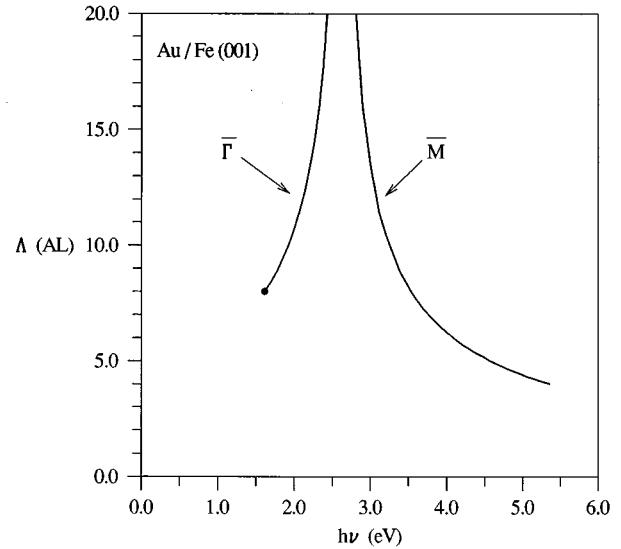


FIG. 6. Period of Kerr effect oscillations for Au (001) overlayers on Fe (001) versus photon energy.

requirements; stationary vectors may also be found on high-symmetry lines or, accidentally, at points of the two-dimensional Brillouin zone having no particular symmetry. In the present paper, for simplicity, we shall restrict our discussion to the high-symmetry points of the two-dimensional Brillouin zone; although contributions arising from other regions of the two-dimensional Brillouin zone cannot be excluded *a priori*, the analogy with the problem of oscillatory interlayer exchange coupling suggests that they are much less likely.

Indeed, a careful inspection shows that the only significant contributions for the Au/Fe(001) case originate from the  $\bar{\Gamma}$  and  $\bar{M}$  high symmetry points.

Figure 4 displays the band structure of Au and Fe the normal direction for the center  $\bar{\Gamma}$  of the (001) two-dimensional Brillouin zone. The vertical arrow indicates the optical transition giving rise to an oscillatory Kerr effect in the Au overlayer. The initial state (dashed line) is a degenerate  $5d_{yz, zx}$  band. The final state (heavy solid line) has predominant  $6p_z$  character. Thus, the optical transition has a strong matrix element. The Fe states of symmetry corresponding to the final state of the optical transition are indicated by the heavy solid line. For majority spin electrons, the final state is only weakly confined in the Au layer because a state of appropriate symmetry is available in the Fe substrate; in contrast to this, for minority spin electrons, no states are available in the Fe so that one has complete confinement of the final state of the optical transition. In addition to this, the initial band is rather flat, whereas the final band is steep. Thus, according to the above theory and discussion, the transition shown in Fig. 4 should give rise to a strong oscillatory Kerr effect in the Au overlayer, with a period given by the reciprocal length of the horizontal arrow.

The band structure of Au and Fe for the  $\bar{M}$  point of the (001) two-dimensional Brillouin zone is shown in Fig. 5. Here, the optical transition occurs between a very perfectly flat  $5d_{xy}$  band and steep bands of  $6p_x$  or  $6p_y$  character. Again, this is a transition of strong matrix elements. The final state is very weakly confined for majority spin electrons,

whereas complete confinement is obtained for minority spin electrons, up to about 3.3 eV above the Fermi level. This transition is also expected to give rise to a strong oscillatory Kerr effect in the Au overlayer on Fe (001), with a period given by the horizontal arrow.

The variation of the oscillation period  $\Lambda$  versus photon energy  $h\nu$  is presented in Fig. 6. The solid point indicates the onset of the transition, where the final state reaches the Fermi level. The period exhibits a pronounced variation with photon energy, and in particular, diverges for  $h\nu=2.63$  eV,

where the final state reaches the band edge. Such a characteristic behavior should allow a very clear experimental test of the theory given in the present paper.

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