Theory for the giant magneto-optical Kerr rotation in CeSb

U. Pustogowa, W. Hübner, and K. H. Bennemann

Institute for Theoretical Physics, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

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Abstract

We calculate the linear magneto-optical Kerr rotation angle for CeSb in the near-infrared spectral range. Using an exact formula for large Kerr rotation angles and a simplified electronic structure of CeSb we find at $\hbar\omega = 0.46$ eV a Kerr rotation of $90^\circ$ which then for decreasing $\omega$ jumps to $-90^\circ$ in very good agreement with recent experimental observations. We identify the general origin of possible $90^\circ$ polarization rotations from mainly optical properties and discuss its relation to the magnetic moments and magnetic dichroism of the material.

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The search for large Kerr rotations in magneto-optics has been a longstanding subject of both basic and application-oriented research. The prediction and observation of giant Kerr rotations [1–3] in the nonlinear magneto-optical Kerr effect (NOLIMOKE) on multilayer sandwiches and thin magnetic films have successfully demonstrated the enhanced sensitivity of nonlinear optics to magnetism, in particular on low-dimensional systems due to the reduction of symmetry. Although the detailed values of the enhanced Kerr rotation in NOLIMOKE depend on the electronic structure of the investigated system, the spin-orbit coupling strength, the magnetization direction in the sample and the polarization of the incoming light, the $90^\circ$ nonlinear Kerr rotation in the longitudinal configuration and steep angle of incidence [3], is essentially independent of the frequency and results from symmetry considerations.

Prior and also parallel to NOLIMOKE, the search for enhanced Kerr rotations has been pursued in the conventional linear magnet-optic Kerr effect (MOKE). Since the MOKE rotation of transition metals like Fe or Ni is typically in the range of only some $0.1^\circ$ for optical frequencies one had to resort to particular magnetic rare-earth or uranium alloys with large spin-orbit coupling constants, large magnetic moments, and with special optical resonances in only one spin type to find relatively large Kerr rotations at low temperatures ($\approx 1 \text{ K}$) and at lower frequencies. Furthermore, the application of large external magnetic fields ($\approx 5 \text{ T}$) was necessary. In this way, a record-high MOKE rotation of $14^\circ$ has been observed for CeSb in 1986 by Schoenes et al. [5]. Very recently, however, the same group (Pittini et al. [6]) observed the largest observable rotation of $90^\circ$ and an abrupt jump of this rotation to -90$^\circ$ in CeSb by reducing the frequency from 0.55 eV to 0.46 eV.

In this paper, we present a simplified model for such a giant MOKE rotation. We argue that this large Kerr rotation occurs for a frequency set by both the plasma frequency and special optical transitions allowed only in one spin type, rather than the magnetic properties. Our explanation relies largely on an improved evaluation of the Kerr rotation for a model band structure which does not assume any small parameters in the off-diagonal components of the reflected electric field. We do not need all details of the complicated Ce monopnictides bandstructure to explain the large Kerr angle. In CeSb the effect of the Ce $f$ electrons is merely to spin-polarize and split up the Sb $p$ bands via hybridization. The simplified theory
outlined in the following describes however already the physical origin of the large linear Kerr effect.

The relation of MOKE signals (Kerr rotations or intensitiy measurements) to magnetic properties of the material is of special interest. Thus, MOKE is used for the determination of the direction and relative strength of the magnetization in saturated ferromagnetic materials. We show hereby analysing the reflectivity, the magnetic dichroism and the electrical-susceptibility-tensor elements that this special 90° Kerr-angle resonance is based on the behavior of the diagonal (‘nonmagnetic’) susceptibility $\chi_{xx}(\omega)$ and thus is not related to the magnitude of the magnetization in the sample.

To determine large Kerr rotation angles we have to use general expressions for the Kerr rotation and ellipticities in contrast to the usually taken linearized formulae which are valid if $\tan \varphi \approx \varphi$ holds. Following general ellipsometry arguments the complex polar Kerr angle $\kappa$ is defined as

$$\kappa = \frac{E_y}{E_x} = \frac{\tan \varphi + i \tan \varepsilon}{1 - i \tan \varphi \tan \varepsilon},$$

where, $\varphi$ is the Kerr rotation angle and $\varepsilon$ is the ellipticity. $E_y$ and $E_x$ are components of the reflected electric field. Solving Eq. (1) we find for the Kerr angle

$$\varphi = \arctan \left( \frac{-1 - |\kappa|^2}{2 \text{Re}(\kappa)} \pm \sqrt{\left(1 - |\kappa|^2\right)^2 + 4 \left[\text{Re}(\kappa)\right]^2} \right)^{1/2} + \varphi_0.$$  

(2)

The analysis of Eq. (2) yields that $180^\circ$-jumps in $\varphi$ may occur when $\text{Re}(\kappa)$ is zero or infinity. The behavior of $\kappa$ results from the well known expression for the linear polar Kerr rotation

$$\kappa = -\frac{\chi^{(1)}_{xy}(\omega)}{\chi^{(1)}_{xx}(\omega)} \sqrt{1 + \chi^{(1)}_{xx}(\omega)}.$$  

(3)

Here, $\chi^{(1)}_{xx}$ and $\chi^{(1)}_{xy}$ denote the elements of the linear susceptibility tensor. The complex value $\kappa$ describes the tangent of the angle, $\kappa = \tan \Phi_K$, with $\Phi_K = \varphi + i \varepsilon$. Note, for small Kerr rotations Eq. (2) reduces to $\varphi = \text{Re}(\kappa)$. Eq. (2) can be rewritten as

$$\varphi = \frac{1}{2} \arctan \left( \frac{2 \text{Re}(\kappa)}{1 - |\kappa|^2} \right) + \varphi_0,$$

(4)

with $\varphi_0 = 0$ for $|\kappa|^2 \leq 1$, $\varphi_0 = 90^\circ$ for $|\kappa|^2 > 1$, $\text{Re}(\kappa) \geq 0$, and $\varphi_0 = -90^\circ$ for $|\kappa|^2 > 1$, $\text{Re}(\kappa) < 0$. For details see Groot Koerkamp [7,8]. Thus, the behaviour of $\varphi$ for $\text{Re}(\kappa) \to 0$
depends on the value of $|\kappa|^2$. For $|\kappa|^2 \leq 1$, the case of small Kerr rotations, together with $Re(\kappa)$ also $\varphi$ goes to zero. In the other case, for $|\kappa|^2 > 1$, changing the sign of $Re(\kappa)$ yields to different values of $\varphi_0$, including a jump from $90^\circ$ to $-90^\circ$. So far, Kerr effect measurements realized only the first $|\kappa|^2 \leq 1$ case and no $\pm 90^\circ$ jump was observed. The analytical analysis of Eq. (3) yields that $Re(\kappa)$ can get zero for $Re(\chi_{xx}) = 0$. Thus, we define two conditions for the occurence of a $\pm 90^\circ$ jump in $\varphi$

$$|Im(\kappa)| \geq 1 \quad \text{and} \quad Re(\chi_{xx}) = 0$$  \hspace{1cm} (5)

The ellipticity angle $\varepsilon$ is given by

$$\varepsilon = \frac{1}{2} \arcsin \left( \frac{2Im(\kappa)}{1 + |\kappa|^2} \right).$$  \hspace{1cm} (6)

Note, these results are of general interest, in particular the occurence of a "resonance" behaviour in the Kerr rotation.

For describing now the infrared Kerr-spectrum of CeSb as measured by Pittini et al. [6] we use only one feature of the electronic structure of CeSb close to the Fermi level, namely fairly flat and nearly parallel bands above and below the Fermi level $\varepsilon_F$ in the $\Gamma-Z$ direction [9]. The $p$ states of Sb are split by approximately 0.6 eV. Furthermore, via hybridization with the $f$-electron minority-spin subband of CeSb (lying $\sim$ 3 eV above $\varepsilon_F$) the Sb $p$ states have an induced spin polarization. Thus, there are favored transitions of one spin sort between the $p$ states of very high weight [10]. In this case, the interband parts of the diagonal and off-diagonal susceptibilities can be written as sums over transitions for one spin band only. Thus,

$$\chi_{xx} = \chi_{xx,\text{intra}} + \sum_{l,l'} L_{l,l'}, \quad \chi_{xy} = -\frac{\lambda_{s.o.}}{\hbar\omega} \sum_{l,l'} L_{l,l'}, \quad \chi_{yy} = \frac{\lambda_{s.o.}}{\hbar\omega} \sum_{l,l'} L_{l,l'},$$  \hspace{1cm} (7)

where $\chi_{xx,\text{intra}}$ denotes the intraband contribution important only in the diagonal elements of $\chi$, $L_{l,l'}$ denotes the response from transitions between the bands $l$ and $l'$ and $\lambda_{s.o.}$ is the spin-orbit coupling constant. Note, the factor $\lambda_{s.o.}/\hbar\omega$ results from including spin-orbit coupling to lowest order in the wave functions. However, this will not directly effect $\varphi$. The Kerr rotation is then calculated by using

$$\kappa = \frac{\lambda_{s.o.}}{\hbar\omega} \frac{1}{\left(1 + \frac{\chi_{xx,\text{intra}}}{L}\right) \sqrt{1 + \chi_{xx}^{(1)}(\omega)}}.$$  \hspace{1cm} (8)
For simplicity, we approximate the transitions between the (Sb) \( p \) bands by a single atomic (nondispersive) Lorentzian

\[
\sum_{l,l'} L_{l,l'} = L := \frac{1}{E_f - E_i - \hbar \omega + i\hbar \alpha},
\]

with the band positions \( E_f \) of the unoccupied final state and \( E_i \) of the occupied initial state and the damping factor \( \alpha \). Note, optical transitions between minority-spin electrons in these states strongly prevail. Furthermore, we describe intraband effects by a conventional Drude term

\[
\chi_{xx,\text{intra}} = -\frac{\omega_{pl}^2}{\omega(\omega + i\tau)},
\]

with the plasma frequency \( \omega_{pl} \) and the damping \( \tau \). Using these expressions one can then determine \( \varphi \) with the help of Eq. (4).

For a further analysis and for comparison with experiment we also calculate the optical reflectivity \( R \) using

\[
R = \left| \frac{N - 1}{N + 1} \right|^2,
\]

with the refraction index \( N^2 = \epsilon_0 + i\epsilon_1 = 1 + \chi_{xx} + i\chi_{xy} \).

We now present results for the Kerr rotation, the reflectivity and in particular the dependence of the large Kerr angle on the plasma frequency \( \omega_{pl} \) and interband splitting \( \Delta E = E_f - E_i \) referring to the dominant optical transition.

In Fig. 1 the frequency dependence of the linear polar Kerr angle \( \varphi \) and the ellipticity \( \varepsilon \) are shown. Here \( \varphi \) and \( \varepsilon \) are calculated using Eqs. (4), (6), and (8). Parameters are choosen such as to obtain the jump in \( \varphi \) from -90° to +90° at precisely 0.46 eV as measured by Pittini et al. [6]. Thus, we use for the interband splitting \( \Delta E = 0.67 \) eV in agreement with the bandstructure of Liechtenstein et al. [9] and for the damping \( \alpha = 0.1 \) eV as usually taken for Kerr angle calculations. In the Drude term the parameters \( \omega_{pl} = 0.93 \) eV and \( \tau = 0.95 \times 10^{-4} \) eV are used. The value for \( \tau \) is taken from Kwon et al. [11]. Note, the jump at 0.46 eV is really a jump from an angle -90° to an angle +90° without intermediate values. The ellipticity angle changes sign at 0.41 eV going from a minimum of -20° to the maximum of 35° . For comparison the experimental values by Pittini et al. [6] are shown as dots.
In Fig. 2 we show furthermore the frequency dependence of the optical reflectivity $R(\hbar \omega)$ in the energy range from 0 to 0.8 eV. For the Kerr angle spectrum the most important range of this curve is the deep minimum at 0.46 eV, at the same energy where the jump in $\varphi$ occurs. The discrepancy with respect to experimental results is presumably due to our simplified electronic structure of CeSb since we neglect interband transitions at large frequencies.

Figs. 3 and 4 show the dependence of $\varphi(\hbar \omega)$ and, in particular, of the jump position on $\Delta E$ and the plasma frequency $\omega_{pl}$. Fig. 3 shows Kerr-angle spectra $\varphi(\hbar \omega)$ with plasma frequencies varying from 0.2 eV to 1.3 eV. Note, for plasma frequencies higher than a threshold value depending on $\Delta E$ the 180°-jump vanishes, whereas for low plasma frequencies the jump amplitude is stable, while only the position of the jump moves to lower energies. The variation of the interband transition $\Delta E$ yields the opposite behavior, see Fig. 4. Here, $\Delta E$ changes from 0.2 eV to 1.6 eV. The jump in $\varphi$ vanishes for low $\Delta E$ and moves for large values of $\Delta E$ to higher energies. Thus, we find that the ratio of the plasma frequency $\omega_{pl}$ to $\Delta E$, which characterizes the interband transitions, is essential for the occurrence of a 180° jump in the Kerr angle.

The position of the 180° jump in the Kerr angle $\varphi$ is determined by a zero of the real part of $\chi_{xx}$, in particular $(\chi_{xx,intra} + L) = 0$. This equation yields a condition for the ratio of $\omega_{pl}/\Delta E$ at which the Kerr-angle jump may occur. Note, this equation reflects the ratio $\chi_{xx}/\chi_{xy}$ using two approximations, namely (i) $\chi_{xy} \sim \lambda_{so}/\hbar \omega$ and (ii) assuming interband transitions only in one spin subband. Approximately, for $\omega_{pl}/\Delta E \leq 1.5$ the Kerr angle jump disappears. Moreover, there is no lower boundary for the occurrence of this jump as a function of the ratio $\omega_{pl}/\Delta E$.

For all parameter sets we analyzed the realization of the jump conditions Eq.(4). The second one, $Re(\chi_{xx}) = 0$ was fulfilled in every case indicating a resonance in $\varphi$. The additional analysis of the value of $|\kappa|$ or, at the resonance of $Im(\kappa)$, enable us to distinguish between less exciting and already discussed resonances for $|Im(\kappa)| < 1$ and the ’degenerated’ resonances with the ±90° jump in $\varphi$ for $Im(\kappa) \geq 1$.

Note, the jump in the Kerr rotation can be described also by a continuous rotation from 0° to 180°, which are identical polarization angles. Then, the condition $Im(\kappa) = 1$ indicates the boundary between those resonances, which return the angle to zero and in the other case
those, which allow the continuous rotation. The jump occurs only as a result of implementing
the usual boundary conditions: \( \varphi = 0 \) Kerr rotations outside the 'excited' frequency region.

In the energy range below \( \approx 0.5 \text{ eV} \) our results for the Kerr angle \( \varphi(\hbar\omega) \) and Kerr
ellipticity \( \varepsilon(\hbar\omega) \) yield good agreement with the experimental data. The deviation for larger
energies results from the neglect of further interband transitions. Note, we have included only
the interband transition with \( \Delta E \) in our model. For the same reason as already mentioned
our calculation yields a too large reflectivity at 0.8 eV. The inclusion of more interband
transitions, especially of transitions of the other spin, would decrease the reflectivity in this
energy range and yield results for the Kerr rotation, ellipticity and reflectivity in better
agreement with experiment for larger energies.

Nevertheless, for the explanation of the Kerr angle jump it is not primarily important
to know the exact electronic structure, but to fulfill the resonance condition combining the
important features of the electronic structure with the plasma frequency. This concludes
then the general model for large Kerr rotations due to the algebraic structure of \( \kappa \) and \( \varphi \),
see Eqs. (1,2,3).

Historically, large linear Kerr rotations have been expected in materials with large spin-
orbit coupling like in CeSb. In our model the spin-orbit coupling strength does of course
influence the Kerr angle, namely a large spin-orbit coupling is necessary to realize \( \text{Im}(\kappa) \geq 1 \),
but not influences the jump position managed by \( \text{Re}(\chi_{xx}) = 0 \). In addition, in the case
of CeSb the interband splitting \( \Delta E \) is affected by the spin-orbit coupling. Note, for the
Kerr effect in transition metals the spin-orbit interaction does not appreciably influence the
electronic bandstructure, but of course has to be included in the wave functions.

In view of our model, related materials with pronounced interband transitions due to flat
\( f \) and \( d \) bands could exhibit similar behavior. However, due to the resonance character of
the \( \varphi \) enhancement and the Kerr-angle jump, slight changes of the parameters and dominant
optical transitions might already suppress the giant Kerr effect [14]. This may explain why
for example CeBi with similar \( \lambda_{s.o.} \) and magnetic moments exhibits only a Kerr angle of
about \(-9^\circ \) [15]. The different shapes of \( \varphi(\omega) \) for CeSb and CeBi might suggest already
differences in the dominant dipole transitions and that in CeBi in contrast to CeSb the
resonance does not occur (see \( \varphi \simeq -9^\circ \longrightarrow \varphi \sim 3^\circ \) for \( \hbar\omega = 0.35 \text{ eV} \) to 0.5 eV).
Mostly interesting is the relation of the discussed Kerr-rotation jump to the magnetic properties of the material. In view of this we calculate the magnetic dichroism in MOKE. With reference to the magnetic dichroism in circularly polarized light we define the dichroism for linearly incident polarization

\[ d = \frac{I(+M) - I(-M)}{I(+M) + I(-M)}, \tag{11} \]

with \( I(\pm M) \) the intensities for inversed magnetization directions. Using for the intensities of the reflected light \( I = |E_{refl}|^2 = |\chi \times E_{incident}|^2 \) we find \( I(\pm M) \sim |\chi_{xx} \pm i\chi_{xy}|^2 \) and the dichroism

\[ d = 2\frac{Im(\chi_{xx})Re(\chi_{xy}) - Re(\chi_{xx})Im(\chi_{xy})}{|\chi_{xx}|^2 + |\chi_{xy}|^2}. \tag{12} \]

In Fig. 5 we illustrate the different frequency dependences of the magnetic dichroism \( d \) and the absorptive part of the magnetic susceptibility tensor element \( Im(\chi_{xy}) \). We find a strong dichroism of more than 70% at the \( \varphi \)-jump frequency whereas the spin-polarized absorption depends only on the electronic structure. The maximum in \( Im(\chi_{xy}) \) occurs at \( \Delta E = 0.67 \text{ eV} \). In previous more detailed microscopic calculations of \( \chi_{xy} \) \[16\] we found a linear dependence of amplitudes of pronounced maxima in \( \chi_{xy} \) on the magnetic moment and this seems to be a general property. On the other hand, inspecting Eq.( 3) (small Kerr effect) for a weak varying denominator we find \( \varphi \sim \chi_{xy} \sim M \). For the \( \varphi \)-jump case, in contrast, the Kerr rotation and the dichroism are determined by nonmagnetic optical properties (\( Re(\chi_{xx}) = 0 \)) not resolving the magnetism of the material. Thus, we analyzed a case of polarization rotation in a magnetic material not caused by magnetic properties and, strictly speaking, cannot register this effect as magneto-optical Kerr effect. Experimentally, the usual MOKE and the here discussed polarization rotation can be distinguished by an additional analysis of the optical reflectivity.

It would be of interest to determine also the nonlinear Kerr rotation in CeSb. Generally, the nonlinear Kerr effect involving more transitions will be a more sensitive probe of the electronic structure. Also, the nonlinear Kerr rotation would illustrate the different nature of giant Kerr rotations in linear and nonlinear optics. The theoretical analysis for this follows from previous studies \[1,3\].
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\[ \varphi = \frac{1}{2} \arctan \left( \frac{2Re(\kappa)}{1 - |\kappa|^2} \right) . \]

The solution of Eq. (1) for \( \varphi \) and \( \varepsilon \) starts with rewriting this complex equation into two real equations for \( Re(\kappa) \) and \( Im(\kappa) \). Whereas Eq. (2) was found by straight-forward substitution of \( \tan \varepsilon \), another solution is found adding \( |Re(\kappa)|^2 \) and \( |Im(\kappa)|^2 \) and subsequent using the addition theorem between \( \tan \varphi \) and \( \tan 2\varphi \). Than, the following inversion of \( \tan 2\varphi \) yields to an unphysical halving of the definition range to \( [-\pi/4, \pi/4] \), which is compensated by the angle \( \varphi_0 \) extending the definition range to \( [-\pi/2, \pi/2] \).

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Note, these conditions change when transitions in both majority and minority spin bands have to be considered.

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Note the differences between CeSb and CeBi regarding the ellipticity and the off-diagonal conductivity $\sigma_{xy}$, s. Pittini et al.. Note, CeBi exhibits less structure in $\sigma_{1xy}$ and $\sigma_{2xy}$ and a different frequency dependence suggesting different role of important electric dipole transitions.

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FIGURES

FIG. 1. Calculated frequency dependence of the linear magneto-optical Kerr-angle $\varphi$ (solid curve) and the ellipticity angle $\varepsilon$ (dashed curve) of CeSb calculated with a Lorentz-type susceptibility. For comparison also experimental results of Pittini et al. for $\varphi(\hbar\omega)$ and $\varepsilon(\hbar\omega)$ are shown.

FIG. 2. Optical reflectivity $R$ of CeSb calculated from the diagonal susceptibility $\chi_{xx}(\omega)$. Experimental values are also shown.

FIG. 3. Dependence of the Kerr angle spectra $\varphi(\hbar\omega)$ of CeSb on the value of the plasma frequency $\omega_{pl}$. For every curve the value of $\omega_{pl}$ (in eV) is shown.

FIG. 4. Dependence of the Kerr angle spectra $\varphi(\hbar\omega)$ on the interband transition energy $\Delta E$. For every curve the corresponding value of $\Delta E$ (in eV) is shown.

FIG. 5. Frequency dependence of the magnetic dichroism $d$ and the absorptive part of the magnetic susceptibility $\text{Im}(\chi_{xy})$. 
Fig. 1

Kerr rotation angle $\phi$ and ellipticity $\epsilon$ (deg)

CeSb

Exp. Pittini: $\triangle \phi$

$\square \epsilon$

energy (eV)

0.2 0.4 0.6 0.8

-90.0 -60.0 -30.0 0.0 30.0 60.0 90.0

$\epsilon$

$\phi$
Fig. 3

CeSb

variation of $\omega_{pl}$ (in eV)

$\lambda_{s.o.} = 0.5$ eV
CeSb

$\lambda_{\text{s.o.}} = 0.5 \text{ eV}$
Fig. 5

magnetic dichroism $d$

$\epsilon$

$\varphi$

$\Delta E$

energy (eV)

$\text{Im} \chi_{xy}$