Discovery of 90 degree Magneto-optical Polar Kerr Rotation in CeSb

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We report the first discovery of 90° magneto-optical Kerr rotation in CeSb at 1.5 K in a field of 5 T. This is 6.5 times more than the previous record rotation of 14° and it is the absolute maximum observable rotation in a single reflection. Using newly derived equations for large rotations and ellipticities, the off-diagonal conductivity has been computed from the magneto-optical spectra and the optical spectra determined for the same external parameters. The spectra are discussed in terms of the field and temperature dependent ground state of CeSb. [S0031-9007(96)00717-X]

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CeSb is, together with CeBi, one of the most fascinating rare-earth compounds owing to its variety of anomalous electronic and magnetic properties which contrast the simplicity of its rocksalt structure. In particular, many features connected with the $4f^1$ state are still unexplained. The resonant photoemission spectra of the cerium monopnictides show a f-related double peak [1], for which several different models have been proposed [2–6]. Furthermore, the crystal-field splitting of the $4f^1$ state of the cerium monopnictides is strongly reduced [7] by f-band mixing effects. In the magnetically ordered phase, CeSb and CeBi show a very strong magnetic anisotropy, which cannot be explained with the symmetry of the $4f^1$ ground state Γ_7. CeSb is the material with the most complicated and intriguing magnetic phase diagram among the cerium monopnictides. Last but not least, the largest known magneto-optical Kerr rotation angle of 14° was measured in CeSb [8].

The occurrence of very large magneto-optical Kerr rotations in materials on the borderline between well-localized and itinerant f electron states has recently attracted much interest from the theoretical side. Different approaches have been chosen to tackle the difficult problem to compute magneto-optical spectra of uranium and cerium compounds, like US [9–11], USe, UTe [10,11], CeTe [11], and CeSb [11–13]. The until now largest rotation of 14°, reported for the latter compound, made this material become a favored object of investigation. Yet the low-energy limit of 0.55 eV of the magneto-optical spectrometer used by Reim *et al.* [8] allowed us only to see a strong increase of the Kerr rotation with decreasing photon energy, while the maximum was not reached. We have now extended the low-energy limit of the Kerr effect measurements to 0.23 eV [14] and indeed found a maximum of 90° single Kerr rotation at 0.46 eV. This is one order of magnitude larger than the maximum Kerr rotation in any other material. Present band theoretical treatments have predicted rotations of only a few degrees [9–13]. Therefore, the discovery of 90° rotation will ask for new concepts to describe the exact nature of the $4f$ state in this compound and its coupling to the d and p states of Ce and Sb, respectively. In addition, the great variation of the spectrum as function of the magnetization indicates that magnetic quantum numbers play an important role. It is to be expected that a suitable theory has to include a Coulomb correlation energy which depends on these quantum numbers. Also the unprecedented high Kerr rotation and ellipticity values made it necessary to derive new expressions for relating the Kerr spectra to the off-diagonal conductivity, as the commonly used expressions rely on approximations for small Kerr angles only.

We have measured the infrared magneto-opical Kerr spectrum on single crystals of CeSb at a temperature of 1.5 K with applied fields of 3 and 5 T. In the measurement at 5 T, the magnetic moment of the $4f^1$ state of CeSb reaches the value of $2.06\mu_B$ [15], which is close to the magnetic moment of the free Ce^{3+} ion, $2.14\mu_B$. Therefore we can assume a ground state $|j = \frac{5}{2}, j_z = -\frac{5}{2}\rangle$ for the magnetically saturated $4f^1$ level. The results of our measurements of the saturated infrared Kerr rotation and ellipticity are shown in Fig. 1. Compared with the first results of Reim *et al.* [8] some differences can be recognized in the overlapping spectral range. They arise from the significantly improved resolution on the energy and rotation scales in this infrared region. At the spectral limit of the magneto-optical measurements of 0.55 eV of Reim *et al.* [8], the monochromator slit was completely open. The corresponding low-energy resolution led to a smearing out of the features in the Kerr rotation and ellipticity. Going from higher energies towards the infrared region (Fig. 1), the Kerr rotation θ_K reaches a first minimum of −13.2° at 0.72 eV, which is similar to the −14° observed by Reim *et al.* [8]. At lower photon energies, θ_K crosses the zero line at 0.65 eV and increases reaching first a shoulder of 37.4° at 0.6°. Then at 0.46 eV it reaches the huge value of 90°. A rotation of the polarization direction of 90° is equivalent to a polarization rotation of −90° since both describe the

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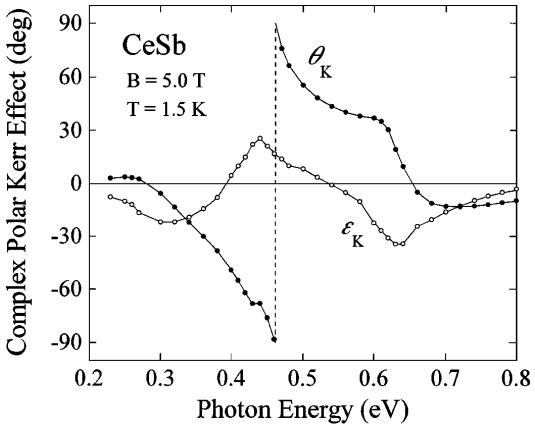

FIG. 1. Complex polar Kerr effect of CeSb measured at $T = 1.5$ K and $B = 5$ T.

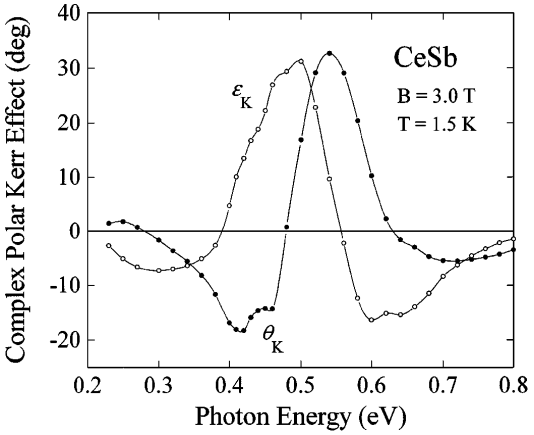

FIG. 2. Complex polar Kerr effect of CeSb measured at $T = 1.5$ K and $B = 3$ T.

same direction of vibration of the electrical field vector. A further increase of the Kerr rotation above 90°, say to 100°, is indistinguishable in a single measurement or spectrum from a rise of the rotation from −90° towards less negative values, i.e., −80° in our example. Thus the maximum absolute value to be detected in a single reflection is 90°. To decide whether the intrinsic signal exceeds 90°, additional measurements for lower spin polarization or magnetization have to be performed. We checked carefully by measuring the field and temperature dependence of the rotation in the neighborhood of the peak energy that the rotation indeed jumps from +90° to −90°. The observed 90° rotation in saturated CeSb should be compared with typical values of other materials. The second largest rotation in the optical range has been observed in pseudobinary $Usb_{0.8}Te_{0.2}$ and amounts to 9° [16]. Fe has a maximum Kerr rotation of −0.53° and the amorphous transition element–rare earth films used for magneto-optical recording display rotations of 0.2° to 0.3° [17].

The energy dependence of the Kerr ellipticity ε_K compared with the Kerr rotation θ_K appears unusual for photon energies between 0.4 and 0.6 eV. Generally, the shape of the Kerr ellipticity is similar to the derivative of the Kerr rotation, as is the case for example in CeSe [18] or also in CeSb if the magnetization is sufficiently reduced and, therefore, the magneto-optical signals are smaller. This manifests the failure of the Kramers-Kronig relation between θ_K and ε_K. Also, as we will show after the derivation of the exact relations between θ_K, ε_K, on one hand, and the real and imaginary parts of the off-diagonal conductivity σ_{1xy} and σ_{2xy}, on the other hand, the ellipticity can never reach 45°.

At the lower field of 3 T, a ferrimagnetic phase with spin alignment ++− is realized in CeSb [19]. The magnetic moment takes the value of one-third of the saturated moment. At 1.5 K and 3 T, a main maximum of the Kerr ellipticity (Fig. 2) appears at 0.5 eV with the value of 31.2°, which is larger than the positive peak of 25.4° observed at 0.44 eV in the field of 5 T. In contrast, the minima of ε_K at 0.31 eV and 0.63 eV are normally reduced from the values observed at 5 T following the trend of the magnetization. The Kerr rotation at 1.5 K and 3 T shows a remarkable main maximum of 32.6° at 0.54 eV. The zeros of θ_K are at almost the same position in both measurements, but the zero at 0.46 eV is a discontinuity in the θ_K curve observed at 5 T while $\theta_K(\hbar\omega)$ is less steep in the measurement at 3 T.

In order to calculate the off-diagonal conductivity with the observed magneto-optical Kerr spectra, we measured the optical reflectivity spectra between 0.01 and 12.4 eV. In the infrared region, we measured simultaneously the Kerr effect and the optical reflectivity of the sample against a gold reference. Therefore the optical constants have been determined from experiments performed under the same conditions of temperature and magnetic field as the Kerr experiment. In Fig. 3, the infrared reflectivity spectra are displayed. Below 0.4 eV, the intraband absorptions, characteristic of the semimetallic behavior of

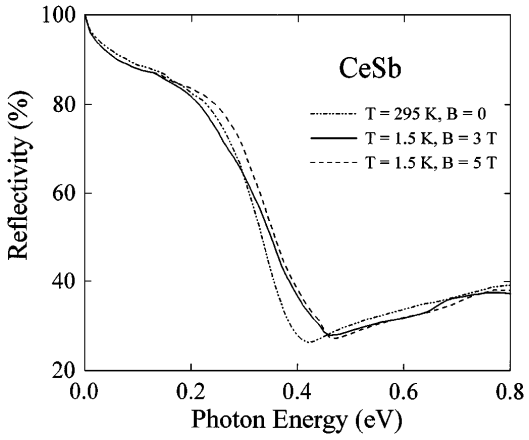

FIG. 3. Near normal incidence optical reflectivity of CeSb measured at room temperature and for the same external parameters as for the magneto-optical measurements.

CeSb, lead to a high reflectivity. At 1.5 K and fields of 3 and 5 T, the plasma minimum has shifted by 0.05 eV to higher energy, as compared to room temperature. The curve at 3 T is peculiar because the effect of the ferrimagnetic ordering leads to a lowering of the reflectivity in the region between 0.2 and 0.3 eV and to the appearance of some structures. In order to obtain the optical constants from a Kramers-Kronig transformation of the optical reflectivity, the reflectivity curves have been extrapolated into the infrared using the Hagen-Rubens law according to the dc resistivity, and beyond 12.4 eV using a ω^{-2} power law first and a ω^{-4} law above 20 eV.

The very large magneto-optical signals do not allow one to use the usual approximative expressions which relate the optical and magneto-optical spectra to the off-diagonal conductivity. Instead, the exact relation between θ_K, ε_K, the optical constants n and k, and the complex off-diagonal conductivity $\tilde{\sigma}_{xy}$ have to be derived.

The exact expression for the off-diagonal conductivity follows from the complex circular optical conductivities

$$\tilde{\sigma}_{\pm} = i\,\frac{\omega}{\pi}\,\frac{\tilde{\rho}_{\pm}}{(1-\tilde{\rho}_{\pm})^2} = \tilde{\sigma}_{xx} \pm i\tilde{\sigma}_{xy}\,, \tag{1}$$

where the complex reflection coefficients $\tilde{\rho}_{\pm}$ for the right (+, rcp) and left (−, lcp) circular light polarizations carry the information on the magneto-optical effects under the form of a complex phase, which vanishes in the absence of a sample magnetization and changes the sign when the field direction is reversed. Therefore the reflection coefficients might be written as

$$\tilde{\rho}_{\pm} = \tilde{\rho}\,e^{\mp i(\theta_K - iD_K)} \tag{2}$$

with

$$\tan\varepsilon_K = \tanh D_K\,. \tag{3}$$

The formulation (2) for the circular reflection coefficients is, in its simplicity, the key for the derivation of an exact relation between the off-diagonal conductivity and the magneto-optical Kerr signal. Inserting (2) into the circular optical conductivities (1) one obtains after some algebra [20]

$$\begin{aligned}\sigma_{1xy} &= -\frac{\omega}{\pi}\,[(Bc + Ad)s_1 + (Ac - Bd)s_2]\,,\\ \sigma_{2xy} &= -\frac{\omega}{\pi}\,[(Ac - Bd)s_1 - (Bc + Ad)s_2]\,,\end{aligned} \tag{4}$$

where A and B are defined by

$$\begin{aligned}A &= n^3 - 3nk^2 - n\,,\\ B &= -k^3 + 3n^2k - k\end{aligned} \tag{5}$$

and c and d follow from

$$c - id = \frac{1}{(a+ib)^2} = \frac{a^2-b^2}{(a^2+b^2)^2} - i\,\frac{2ab}{(a^2+b^2)^2} \tag{6}$$

with

$$a + ib = (n^2 - k^2 - 1)c_1 + 2nkc_2 - n^2 + k^2 - 1 + i[(n^2 - k^2 - 1)c_2 - 2nkc_1 + 2nk] \tag{7}$$

and

$$\begin{aligned}c_1 + ic_2 &= \frac{\cos\theta_K\,\cos\varepsilon_K}{\sqrt{\cos 2\varepsilon_K}} + i\,\frac{\sin\theta_K\,\sin\varepsilon_K}{\sqrt{\cos 2\varepsilon_K}}\,,\\ s_1 - is_2 &= \frac{\sin\theta_K\,\cos\varepsilon_K}{\sqrt{\cos 2\varepsilon_K}} - i\,\frac{\cos\theta_K\,\sin\varepsilon_K}{\sqrt{\cos 2\varepsilon_K}}\,.\end{aligned} \tag{8}$$

Equations (4)–(8) represent the exact relation between the complex off-diagonal conductivity and the optical constants, the Kerr rotation and the Kerr ellipticity. They are valid for any magnitude of the measured Kerr effect. For small Kerr rotations and ellipticities, the usual approximative formulas [21]

$$\begin{aligned}\sigma_{1xy} &\approx -\frac{\omega}{4\pi}\,(B\,\theta_K + A\,\varepsilon_K)\,,\\ \sigma_{2xy} &\approx -\frac{\omega}{4\pi}\,(A\,\theta_K - B\,\varepsilon_K)\end{aligned} \tag{9}$$

are recovered from (4)–(8). From (1)–(3) it follows that, at optical frequencies, the Kerr ellipticity *never* reaches the value of ±45°. Indeed, a Kerr ellipticity $\varepsilon_K = \pm 45°$ is equivalent to a Kerr dichroism $D_K = \infty$. An infinite Kerr dichroism requires from the circular reflection coefficients (2) $\tilde{\rho}_+ = 0$ and $\tilde{\rho}_- = \infty$ (or inversely), except when the field-free reflection coefficient $\tilde{\rho}$ itself vanishes.

Using equations (4)–(8), we have determined the off-diagonal conductivity spectra from the magneto-optical measurements performed at 1.5 K in fields of 5 and 3 T. The off-diagonal conductivity spectrum at 5 T [Fig. 4(a)] does not differ very much from the original spectrum of Reim *et al.* [8]. In Fig. 4(a), the intraband contributions vanish in the background of the much stronger interband transitions. In the measurement at 5 T, two main transitions denoted by B and C are observed at 0.54 and 0.66 eV, respectively. The transition B has a paramagnetic line

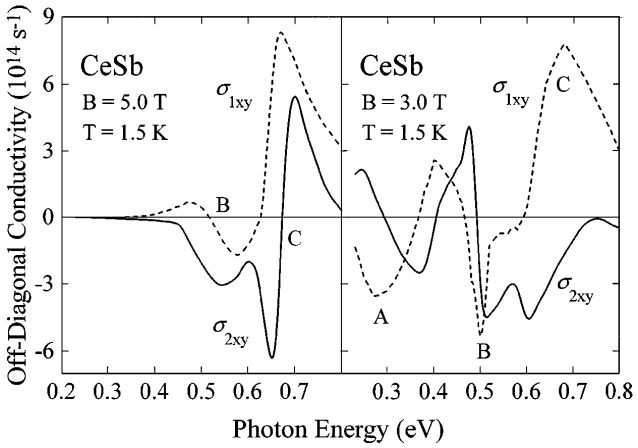

FIG. 4. Off-diagonal conductivity of CeSb, calculated from the magneto-optical spectra using Eqs. (4)–(8).

shape [21] and can therefore unequivocally be assigned to a $4f \rightarrow 5d$ transition for a magnetically saturated $4f^1$ ground state. Indeed, in this case the ground state of the $4f$ level is $|j = \frac{5}{2}, j_z = -\frac{5}{2}\rangle$. Therefore in the $4f \rightarrow 5d$ transition, only the absorption of rcp light, characterized by the selection rule $\Delta j_z = +1$, can occur and in the off-diagonal conductivity no cancellation between the rcp and lcp absorptions takes place. This is one of the reasons for the unconventionally large value of the magneto-optical signal of the $4f \rightarrow 5d$ transition. The corresponding Kerr rotation and ellipticity are further enhanced by the particularly low values of the optical constants in the region of the plasma minimum of the reflectivity. The transition C has a diamagnetic line shape and is assigned to a $5p(\mathrm{Sb}) \rightarrow 5d(\mathrm{Ce})$ transition, where the band states become polarized through f-band mixing effects. At 5 T and 1.5 K, the $4f \rightarrow 5d$ transition can easily be recognized in the off-diagonal conductivity spectrum because it is the only transition with paramagnetic line shape [21]. In their latest calculation of the off-diagonal conductivity of CeSb, Liechtenstein *et al.* and Antropov *et al.* [12] found a small negative peak in σ_{2xy} at 0.3 eV and a larger positive peak in σ_{2xy} at 0.8 eV. An analysis of the matrix elements revealed that the main contribution to these two peaks comes from the transitions from the p bands that strongly interact with the $4f^1$ state.

In the experiment at 3 T [Fig. 4(b)], the $4f \rightarrow 5d$ transition (B) has a diamagnetic line shape, as expected, because several j_z levels mix in the $4f$ ground state in the ferrimagnetic ordered phase. The additional structure A is assigned to the effect of the folding of the Brillouin zone in the ferrimagnetic phase giving rise to the possibility of new interband transitions. The latter are also the origin of the appearance of structures in the reflectivity spectrum at the same energy, noted above.

In conclusion, we have measured for the first time a Kerr rotation of 90° in the infrared magneto-optical spectrum of saturated CeSb. This value of the Kerr rotation is by far larger than all other "giant" Kerr rotations measured before and represents the largest observable rotation in a single reflection. While a quantitative explanation of this stupendous result has to await new theoretical concepts, we suggest on empirical grounds that the origin of this effect is a "paramagnetic" $4f \rightarrow 5d$ transition centered at 0.54 eV, which is enhanced by the vicinity of a strong $5p(\mathrm{Sb}) \rightarrow 5d(\mathrm{Ce})$ transition and by the low values of the optical constants in the region of the plasma minimum. In addition, the magnetic moment of the $4f$ state is saturated under the conditions of the magneto-optical experiment and the $5p$ states become spin polarized through the p-f mixing effect. The strongly anisotropic p-f mixing leads to the "effective" $4f$ level [22], which is found, from the results presented here, to have a binding energy of about 0.54 eV. In the off-diagonal conductivity, the intraband transitions vanish in the background of the $4f \rightarrow 5d$ and $5p \rightarrow 5d$ transitions.

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[1] A. Franciosi, J. Weaver, N. Martensson, and M. Croft, Phys. Rev. B **24**, 3658 (1981).
[2] W. Gudat, M. Iwan, R. Pinchaux, and F. Hulliger, in *Valence Instabilities*, edited by P. Wachter and H. Boppart (North-Holland, Amsterdam, 1982), p. 249.
[3] A. Fujimori, Phys. Rev. B **27**, 3992 (1983).
[4] O. Sakai, H. Takahashi, M. Takeshige, and T. Kasuya, Solid State Commun. **52**, 997 (1984).
[5] W.-D. Schneider, B. Delley, E. Wuillod, J.-M. Imer, and Y. Baer, Phys. Rev. B **32**, 6819 (1985).
[6] M. R. Norman and D. D. Koelling, Phys. Rev. B **33**, 6730 (1986).
[7] R. J. Birgeneau, E. Bucher, J. P. Maita, L. Passell, and K. C. Turberfield, Phys. Rev. B **8**, 5345 (1973).
[8] W. Reim, J. Schoenes, F. Hulliger, and O. Vogt, J. Magn. Magn. Mater. **54–57**, 1401 (1986).
[9] M. S. S. Brooks, T. Gasche, and B. Johansson, J. Phys. Chem. Solids **56**, 1491 (1995).
[10] T. Kraft, P. M. Oppeneer, V. N. Antonov, and H. Eschrig, Phys. Rev. B **52**, 3561 (1995).
[11] S. P. Lim, B. R. Cooper, Q. G. Sheng, and D. L. Price, Physica (Amsterdam) **186B–188B**, 56 (1993); B. R. Cooper, Q. G. Sheng, S. P. Lim, C. Sanchez-Castro, N. Kioussis, and J. M. Wills, J. Magn. Magn. Mater. **108**, 10 (1992).
[12] A. I. Liechtenstein, V. P. Antropov, and B. N. Harmon, Phys. Rev. B **49**, 10 770 (1994); V. P. Antropov, A. I. Liechtenstein, and B. N. Harmon, J. Magn. Magn. Mater. **140–144**, 1161 (1995).
[13] A. Narita and J. Schoenes, Physica (Amsterdam) **186B–188B**, 580 (1993).
[14] R. Pittini, PhD thesis, Eidgenössische Technische Hochschule Zürich, Zürich, Switzerland, 1995.
[15] J. X. Boucherle, A. Delapalme, C. J. Haward, J. Rossat-Mignod, and O. Vogt, Physica (Amsterdam) **102B**, 253 (1980).
[16] W. Reim, J. Schoenes, and P. Wachter, IEEE Trans. Magn. **20**, 1045 (1984).
[17] See, for example, J. Schoenes, in *Electronic and Magnetic Properties of Metals and Ceramics*, edited by K. H. Buschow, Materials Science and Technology Vol. 3A, edited by R. W. Cahn, P. Haasen, and E. J. Kramer (VCH, Weinheim, 1992), p. 147.
[18] R. Pittini, J. Schoenes, and P. Wachter, Physica (Amsterdam) **206B & 207B**, 92 (1995).
[19] J. Rossat-Mignod, P. Burlet, J. Villain, H. Bartholin, T. S. Wang, D. Florence, and O. Vogt, Physica (Amsterdam) **86B–88B**, 129 (1977); Phys. Rev. B **16**, 440 (1977).
[20] R. Pittini and J. Schoenes (to be published).
[21] W. Reim and J. Schoenes, in *Ferromagnetic Materials*, edited by K. H. J. Buschow and E. P. Wohlfarth (North-Holland, Amsterdam, 1990), Vol. 5, p. 133.
[22] H. Takahashi, T. Kasuya, J. Phys. C **18**, 2697 (1985).