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Magneto-optics as a tool to study occupied and empty 4f and 5f states

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Abstract

Magneto-optical spectroscopy is known to be particularly powerful for studying occupied 4f and 5f states through f→d excitations. The recent discovery of d→f transitions in LaSe has revived the idea to use magneto-optical spectroscopy also to study empty f states. We review the state of the art of optical and magneto-optical investigations of occupied and empty f states for lanthanum, cerium and uranium monochalcogenides and monopnictides. © 1998 Elsevier Science S.A.

Keywords: Magneto-optical Kerr effect; Cerium monochalcogenides; Cerium monopnictides; Uranium monochalcogenides; Uranium monopnictides

1. Introduction

Magneto-optical spectroscopy is known for decades to be a powerful tool for the investigation of occupied 4f and 5f states in rare-earth and actinide compounds, respectively [1]. The proportionality of the Faraday and the polar magneto-optical Kerr effect to the product of the spin polarization and the spin–orbit splitting strongly enhances the signal originating from f states compared to that from s and p states [2]. Furthermore for localized f states, the coupling between the single f electrons into a many-electron state [3] and possibly also of the remaining f^{n-1} state with the excited d-electron [2] leads to very structured spectra which give beautiful fingerprints of the f-electron configuration. The reader interested in these aspects of magneto-optical spectroscopy is referred to the cited original work or to a recent review by the author [4]. In the present review, we will limit the discussion of occupied f states to the more delocalized systems, i.e. the $4f^1$ state in cerium compounds and the $5f^3$ state in uranium compounds. This delocalization leads to a larger overlap of the f-wave function with the d-wave function, increasing the matrix element for f→d, as well as for d→f transitions. A beautiful example of this effect of delocalization has been found in CeN [5]. In this compound, often termed intermediate valent, the d→f oscillator strength is so large that d→f and (pd)→f transitions contribute as dominant structures, even in the diagonal element of the optical conductivity. New band-structure calculations [6] corroborate the assignments made already in 1984. In the series of the uranium monochalcogenides, the localization of the f states increases with increasing anion radius, and the diagonal as well as the off-diagonal conductivity display a clear decrease of the intensity of the d→f peak along the series [7]. For the cerium monopnictides with heavier anions, like antimony and bismuth, no d→f transitions have been identified, neither in the diagonal nor the off-diagonal conductivity. Yet, f→d transitions have been found to give rise to Kerr rotation signals up to 90° in CeSb [8]. In the cerium monochalcogenides the situation is similar: d→f transitions are not observed, but f→d transitions lead to Kerr rotations reaching 22° in CeS [9,10]. Finally, the evidence for the discovery of a d→f transition in paramagnetic LaSe [11] will be discussed.

2. Uranium monochalcogenides and monopnictides

The first UX compound (X=S, Se, Te, N, P, As, Sb, Bi) which has been investigated by optical spectroscopy was USb in 1978 [12]. The spectra were interpreted in terms of f→d and p→d transitions. Although evidence for a hybridization between 5f and 6d wave functions was found, the quite localized nature of the $5f^3$ state did not allow the observation of transitions into empty f states. In the following year, US [13] and USe [14] were investigated with the same technique. The spectra showed near 3 eV an additional structures not present in USb. The occurrence of two peaks in the optical conductivity of US and USe below the p→d transition energy range compared to only one transition in USb lead us to assign one of these peaks to a

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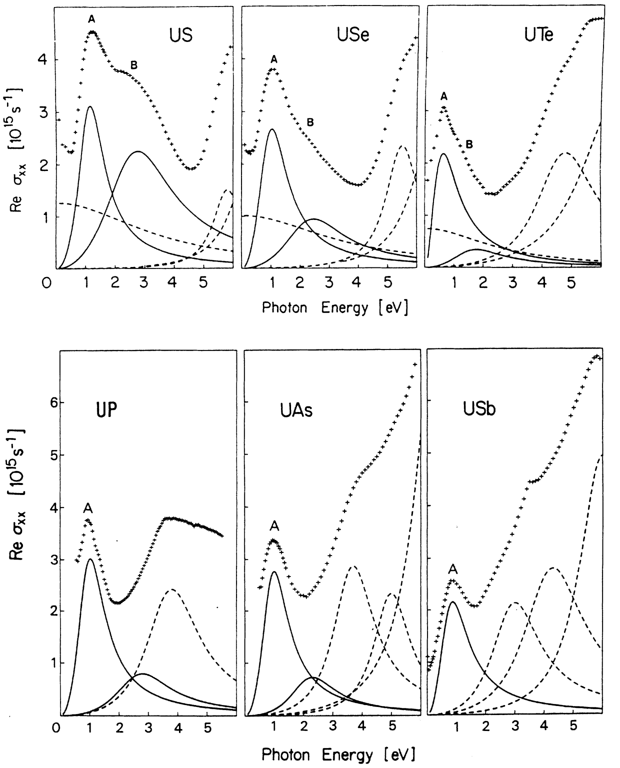

Fig. 1. Lorentz fit of the absorptive part of the off-diagonal conductivity σ_{1xx} for the uranium monochalcogenides and monopnictides.

d→f transition. The results of a molecular orbital calculation suggested [13] that the d→f transition occurs at lower energy than the f→d transition. However, the following systematic investigation of the optical and also magneto-optical properties of the large majority of the uranium monopnictides and monochalcogenides gave increasing empirical evidence for the opposite order, i.e. that the f→d transition occurs at lower energy. Fig. 1 displays the low energy part of the absorptive part of the diagonal conductivity as obtained by a Kramers–Kronig transformation of near-normal incidence reflectivity measurements extending from 0.03 to 12 eV [15] and a decomposition into up to four Lorentzians and one Drude term for US, USe, UTe, UP, UAs, and USb [16,17]. Table 1 lists the parameters for transitions marked A and B in Fig. 1 as a result of the fits [16,17]. Both the figure and the table indicate two trends: if one compares a chalcogenide with the corresponding pnictide (for example US and UP) one notes that peak A has the same oscillator strength, while peak B has a much larger oscillator strength in the chalcogenide; if one compares the compounds within the series of chalcogenides or pnictides, the oscillator strength of peak A is constant, while that of peak B decreases with increasing lattice parameter. The first trend reflects the different concentration of occupied 6d states in the chalcogenides and pnictides. In first approximation, the chalcogenides have one more occupied 6d state than the pnictides. Thus, if the delocalization and hybridization of the 5f states generates 0.2 to 0.5 conduction electrons per formula unit (c.e./f.u.) in the pnictides, one expects 1.2 to 1.5 c.e./f.u. in the corresponding chalcogenide. Assuming for similar lattice parameters similar matrix elements, the oscillator strength for the d→f transition should then be a factor three to six times larger in the chalcogenide than in the corresponding pnictide. The explanation for trend number two is more subtle. With increasing lattice parameter the localization of the 5f state increases and eventually the Coulomb correlation energy exceeds the ‘band width’ of the 5f state. The energy of the 5f state becomes now dependent on its occupation and the 14-fold degeneracy of the itinerant 5f state is lifted leading to a net reduction of the transition probability for d→f transitions. From these trends the assignment of peak A to a f→d transition and of peak B to a d→f transition is quite obvious.

Table 1
Interband transition parameters from the fits in Figure 1. The values are rounded to 0.05

		US	USe	UTe	UP	UAs	USb
f→d	$h\nu_A$ (eV)	1.15	1.0	0.6	1.05	1.0	0.9
	$h\gamma_A$ (eV)	1.1	1.2	1.15	1.1	1.1	1.2
	f_A	0.85	0.9	0.9	0.9	0.9	0.9
d→f	$h\nu_B$ (eV)	2.75	2.45	1.75	2.8	2.3	-
	$h\gamma_B$ (eV)	2.8	2.5	2.1	2.3	1.9	-
	f_B	1.55	0.7	0.25	0.5	0.4	-

The first assignment of a d→f transition in a magneto-optical spectrum was made for US and subsequently also for USe and UTe in 1982 [18,19]. As has been elaborated on with some detail in [20] and in full detail in [16,17], line shape as well as intensity considerations support the assignments already discussed for the optical properties. Fig. 2 displays the Kerr rotation and ellipticity spectra for the three chalcogenides measured at 10 K in an external field of 4 T [19]. Besides the remarkable size of the magneto-optical signal of up to 4°, we note the narrowing of the spectra going from US to USe to UTe. As is most clearly seen in the rotation spectra this narrowing manifests the weakening of one of the two transitions. In a

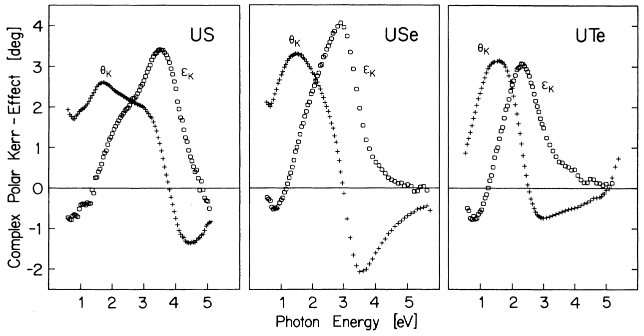

Fig. 2. Polar Kerr-rotation θ_K and ellipticity ϵ_K of the uranium monochalcogenides at T=15 K and B=4 T.

couple of recent publications Oppeneer et al. [21,22] have compared our experimental Kerr spectra with results of relativistic band structure calculations. In the local density approximation (LDA) [21] the agreement between theory and experiment is best for US and worst for UTe. Theory reproduces well the absolute value of the Kerr rotation and ellipticity, as well as the energy of the dominant structure. However, it fails to reproduce the exact shape of the spectra. The LDA calculation predicts two structures of approximately equal weight, the energy separation of which decreases from about 3.5 eV for US to about 2 eV for UTe. In a second computation the LDA is generalized by including on-site Coulomb correlations (LDA+U) [22]. Using for U a value of 2 eV, the absolute value of the Kerr spectra is again well reproduced and the shape has considerably narrowed, so that for UTe a single line results in good agreement with the experiment. On the other hand, the agreement for US is now worse. Thus theory confirms our expectation from the reviewed optical and magneto-optical investigations as well as from transport studies [23,24] that US has rather itinerant 5f electrons which can be reasonably described in an one-particle band-structure calculation, while in UTe the electron correlations are important enough to call for a description in a localized model.

3. Cerium nitride

Among the cerium monochalcogenides and monopnictides (CeX, with X=S, Se, Te, N, P, As, Sb, Bi) cerium nitride plays a particular role. CeN has the most delocalized 4f electron of the series and has been classified as intermediate valent [25,26], while all other CeX compounds exhibit localized 4f behaviour with, however, anomalies of the crystal-field splitting, and in some cases, very rich and peculiar magnetic phases [27–29]. The chemical instability and problems with the stoichiometry of CeN have limited the number of investigations to a very few number. Thus to the best knowledge of the author, magneto-optical measurements have not been performed. Already the optical investigations at room temperature and without an applied magnetic field were very difficult. In 1978, Schlegel et al. [30] published the near-normal incidence reflectivity between 0.03 and 4 eV for single crystals which were polished in high vacuum and then transferred into evacuated spectrometers. The data were analyzed in terms of a Drude model to estimate the free carrier concentration and from this the degree of intermediate valency. In the interband part a shoulder near 1 eV was assigned to a $4f \rightarrow 5dt_{2g}$ transition and a peak at 2 eV was related to a $2p(N) \rightarrow 5dt_{2g}$ transition. In 1984, the present author [5] reported near-normal incidence reflectivity measurements from 0.03 to 12 eV on single crystals which had been cleaved in high and/or ulta-high vacuum in the different spectrometers and which had been measured in-situ. These spectra showed higher reflectivity values and more pronounced structures than the measurements on the polished surfaces. Via a Kramers–Kronig transformation the various optical functions had been derived. The real part of the diagonal conductivity was fitted with six Lorentzians representing in order of increasing energy: intraband ($h\nu=0$ eV), $5d \rightarrow 4f^1$ (1.1 eV), $4f^1 \rightarrow 5d$ (1.93 eV), $(2p5d) \rightarrow 4f^1$ (3.8 eV), and two $2p(N) \rightarrow 5d$ (8.1 and 11.0 eV) transitions. For the first time for a lanthanide compound, these assignments included transitions into empty 4f states. They were done on empirical and theoretical grounds. The empirical arguments were mainly based on a comparison with the UX compounds and on the observation of changes occurring for the spectra on short exposition of the sample to air, i.e. oxidation. The theoretical arguments came from a comparison [5] with results of a APW band-structure calculation of Pickett and Klein [31]. By searching for bands with equal $\mathrm{grad}_{\mathbf{k}}\, E(\mathbf{k})$ over large parts of the Brillouin zone and taking into account the partial densities of states, peaks in the optical conductivity could be related to direct transitions in the band-dispersion scheme [5]. Recently, these assignments have been fully corroborated by a computation of the optical conductivity by Delin et al. [6]. Fig. 3 displays a comparison of the experimental optical conductivity spectrum [5] with the results of computations using the full-potential linear muffin-tin orbital (FPLMTO) method [6]. In one calculation the 4f state is treated as band state, in the other calculation it is treated as core state. Good agreement below 5 eV is only obtained in the former treatment, indicating that the three peaks below 5 eV involve strong f contributions. The analysis of Delin et al. [6] regarding the detailed origin of the peaks in the optical conductivity comes to nearly identical conclusions

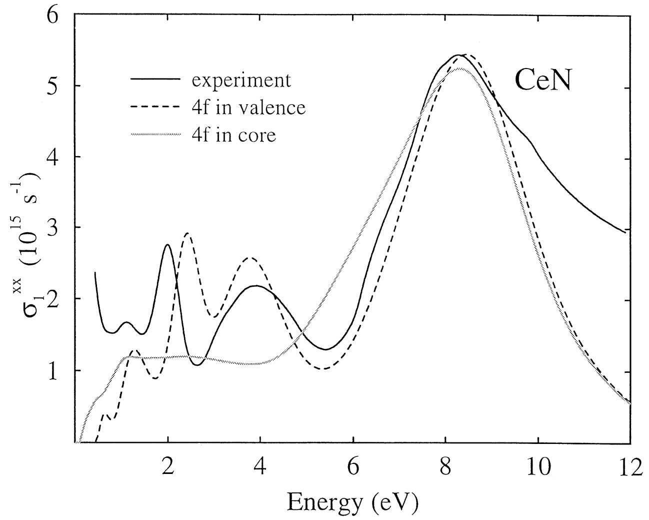

Fig. 3. Comparison of the experimental optical conductivity (full line) of CeN [5] with results of FPLMTO band-structure calculations [6] treating the 4f electrons either as band states (dashed line) or as core states (grey line).

as Schoenes. The peaks near 1 and 2 eV are found to be related to both f→d and d→f transitions. The peak at 4 eV is assigned to transitions originating from p(N) states with strong f and d admixtures to f-dominated final states, and the peak near 8 eV corresponds to p(N)→d transitions. The excellent agreement between experiment and the one-particle band-structure calculation is considered as evidence for an itinerant 4f state in CeN in opposition to an intermediate valent state [6]. We think that CeN is very particular among the 4f and 5f systems, since the two states to be considered for an intermediate valent Ce ion are the $4f^1$ and the $4f^0$ state. So it should not be too surprising, that if these states are pinned at the Fermi energy E_F, a band description of the 4f state gives a much better description than a localized 4f core state situated several eV below E_F.

4. Other cerium monochalcogenides and monopnictides

With the exception of CeN the $4f^1$ state is localized in the cerium monochalcogenides and monopnictides and, contrary to CeN, they all order magnetically. As we have learned from the UX compounds, transitions into localized f states are expected to be weak and if they overlap with strong, electric-dipole allowed transitions they may not be clearly identified. Actually, no d→f nor p→f transitions have been assigned so far to any optical or magneto-optical structure in these CeX compounds. Nevertheless, the magneto-optical properties of the cerium-monochalcogenides and monopnictides are very exciting. In 1986, Reim et al. [32] reported the first magneto-optical investigation on CeX compounds. The authors measured the Kerr-rotation and ellipticity between 0.55 and 5 eV for CeSb, $CeSb_{0.75}Te_{0.25}$, and CeTe at 5 T and 2 K. They observed a shift of the dominant structure from 2 eV in CeTe, to 1 eV in $CeSb_{0.75}Te_{0.25}$, to below the low energy limit of the measurements in CeSb. At 0.55 eV the Kerr rotation of CeSb was found to be 14° which was the largest Kerr rotation reported to this date. The dominant line was assigned to a f→d transition shifting to higher energies with the filling of the conduction band [32]. Pittini and co-workers [8–10] extended the measurements to CeS, CeSe, CeAs and CeBi and added for the cerium monopnictides measurements in the infrared down to 0.23 eV. For CeSb the extension of the measurements to lower photon energies led to the discovery of 90° Kerr rotation at 0.46 eV [8]. In CeBi a maximum rotation of 9.2° was found, while the corresponding rotation peak for CeAs lies still below the new low energy limit of the measurements [9,10]. In the monochalcogenides the largest rotation is 22.1° in CeS [9,10]. The most puzzling question is what causes the huge rotation in CeSb? Is it an intrinsic or an extrinsic effect? It is well known [1] that the size of the rotation is enhanced by small optical constants, i.e. small reflectivity values, and indeed, all rotation maxima in the cerium monochalcogenides and monopnictides occur near the plasma minimum. Yet, this does not necessarily imply that the effect is extrinsic, since it is common that strong interband transitions shift the coupled plasma frequency to near the onset of the fundamental (interband) absorption. A classical example is Ag [33]. Generally, the computation of the off-diagonal conductivity σ_{xy} can help to resolve the problem by comparing the energies of the extrema of $\sigma_{xy}(\omega)$, $\sigma_{xx}(\omega)$ and the Kerr spectra [1]. For the cerium monochalcogenides these agree perfectly, indicating that the Kerr signals are intrinsic. For CeSb the spectra are very rich in structures and small differences of the order of 0.1 to 0.2 eV exist between the extrema in the various spectra. It is not clear at the moment whether this is an indication of some interference enhancement. The reflectivity spectra reported in [8] do not point to an interference effect. On the other hand, preliminary Kerr results obtained in Braunschweig [34] on several other CeSb crystals do not show 90° rotation. It remains to be seen whether different types of Kerr spectra can occur as was found previously for UTe [16,17], or if surface oxidation gives rise to interference effects. In both series of experiments, in Zürich as well as in Braunschweig, the surfaces have been prepared by cleaving the crystals in an inert gas atmosphere and transferring the crystals to the cryostat without exposure to air. To exclude definitively an enhancement of the Kerr-rotation by interference effects one ought to measure the reflectivity before and after the Kerr rotation measurement on the very same samples kept in an inert atmosphere.

Oppeneer et al. [22] have presented arguments for the larger rotation of CeSb compared to CeBi. The analysis of their LDA+U calculation indicates that not only the strong spin and orbitally polarized $4f^1$ level, but also the interaction of this anisotropic $4f^1$ state with the antimony p states contributes to the large signal. Thus, p–f mixing introduced first by Kasuya [35] to account for many of the peculiar properties of cerium monopnictides and subsequently also inferred empirically to account for the large magneto-optical signal [8] seems to be the key property in CeSb.

Besides the large magneto-optical signals for magnetic saturation, CeSb and CeBi show very interesting changes of the shape of the optical and magneto-optical spectra in their various ferri- and antiferrimagnetic phases. In CeBi two and eight additional structures could be observed in the diagonal optical conductivity when the spin arrangement for consecutive (001) planes changes from disorder to $+-+-$ and $++--$, respectively [36]. In CeSb the appearance of new peaks and line shapes in the ferrimagnetic $++-$ phase could be related in detail to a superzone folding of the highest valence band and the lowest conduction band corresponding to a tripling of the unit cell in real space [37].

5. Lanthanum selenide

The lanthanum monochalcogenides have no occupied f state and are Pauli-paramagnets. LaSe becomes superconducting at 1.02 K [38]. Recently Pittini and co-workers [11] have investigated the magneto-optical polar Kerr effect of LaSe at low temperatures and high magnetic fields. Fig. 4 shows the results of the Kerr-effect measurements at 1.5 K in an external field of 10 T, and for comparison, similar data for CeSe. Surprisingly the material with no occupied 4f state displays a Kerr rotation of 2.1°! This is only a factor 2.6 smaller than the Kerr rotation in CeSe. One also notes, that the sharp main lines have very similar shapes, but with reversed sign, and that the energies of the lines are nearly identical. The last fact reflects the similar plasma energies, corresponding to approximately one free electron per formula unit. From the Kerr-rotation and ellipticity spectra and the optical constants, which have been determined from near-normal incidence reflectivity measurements using the Kramers–Kronig relation, the off-diagonal conductivity has been computed. Fig. 5 shows the real (σ_{1xy}) and imaginary part (σ_{2xy}) of the off-diagonal conductivity for the two materials. Again the sharp main lines centered at 2.77 eV in LaSe and 2.60 in CeSe have opposite sign. Both lines have so-called diamagnetic line shape, i.e. the dispersive part of the off-diagonal conductivity σ_{1xy} has the form of an absorption line, while the absorptive part σ_{2xy} has the form of a dispersion curve. LaSe shows a second broader maximum on the high energy side which has its origin in the broader structures found in the Kerr-rotation and ellipticity spectra (Fig. 4) above the sharp line. The shape of σ_{1xy} indicates that this transition is also of diamagnetic line shape and that the overlap of the two lines suppresses in σ_{2xy} the negative oscillation of the high-energy line. A deconvolution gives for the transition energies values of 2.77 and 3.01 eV. The energy difference of 0.24 eV is close to the value of 0.27 eV, known as the energy separation between the $^2F_{5/2}$ and $^2F_{7/2}$ states in trivalent Ce [38].

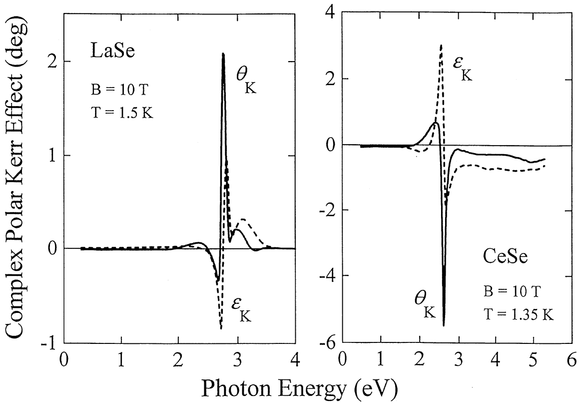

Fig. 4. Polar Kerr-rotation θ_K and ellipticity ϵ_K of LaSe and CeSe in a field of 10 T and temperatures of 1.5 and 1.35 K, respectively.

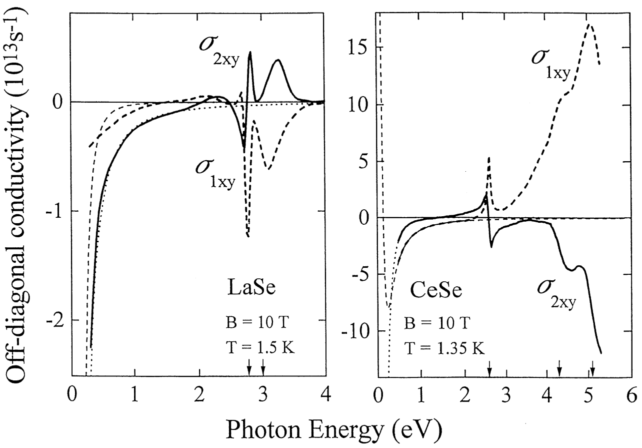

Fig. 5. Dispersive part (σ_{1xy}) and absorptive part (σ_{2xy}) parts of the off-diagonal conductivity of LaSe and CeSe in a field of 10 T and temperatures of 1.5 and 1.35 K, respectively.

The sharp line in CeSe is undoubtedly a $4f^1 \rightarrow 5d$ transition. This transition leads to a diamagnetic line shape when the magnetization is far from saturation [37]. A $5d^1 \rightarrow 4f^1$ transition is expected to have opposite sign since the sign of ΔJ is reversed in the two transitions. Yet, the inversion is not complete since the $4f^1$ state is localized and narrow so that only the $^2F_{5/2}$ ground state will be populated for $k_BT<0.27$ eV, while the $5d^1$ state is broad and the occupied band contains $J=3/2$ as well as $J=5/2$ components. Thus, we expect one diamagnetic transition for the $4f^1 \rightarrow 5d$ transition and two lines for the $5d^1 \rightarrow 4f^1$ transition. This is exactly what we observe in Fig. 5 and gives strong evidence that the magneto-optical signal in LaSe is due to electronic excitations into empty 4f states.

6. Conclusions

We have shown that optical and magneto-optical spectroscopy is not only a powerful tool to study occupied f states, but that in several cases it can also been used very effectively to study empty f states. The most surprising case is certainly represented by LaSe. Although it is not the first time that a substantial magneto-optical signal has been related to a transition into a f state, LaSe represents the first case in which this empty state is a f^0 state and the material has no f moment. The applied magnetic field, however, induces a magnetization and splits the $5d^1 \rightarrow 4f^1$ transition energies for right and left circularly polarized light.

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