Magneto-optical Kerr effect in $Eu_{1-x}Ca_xB_6$

G. Caimi, S. Broderick and H.R. Ott
Laboratorium für Festkörperphysik, ETH Zürich, CH-8093 Zürich, Switzerland

L. Degiorgi
Paul Scherrer Institute, CH-5232 Villigen and Laboratorium für Festkörperphysik, ETH Zürich, CH-8093 Zürich, Switzerland

A.D. Bianchi and Z. Fisk
NHMFL-FSU, Tallahassee FL 32306, U.S.A.
(Dated: March 22, 2022)

PACS numbers: 78.20.Ls, 75.50.Cc

We have measured the magneto-optical Kerr rotation of ferromagnetic $Eu_{1-x}Ca_xB_6$ with $x=0.2$ and 0.4, as well as of YbB$_6$ serving as the non-magnetic reference material. As previously for EuB$_6$, we could identify a feature at 1 eV in the Kerr response which is related with electronic transitions involving the localized 4f electron states. The absence of this feature in the data for YbB$_6$ confirms the relevance of the partially occupied 4f states in shaping the magneto-optical features of Eu-based hexaborides. Disorder by Ca-doping broadens the itinerant charge carrier contribution to the magneto-optical spectra.

Recent investigations of the magneto-optical (MO) properties of EuB$_6$, a ferromagnet with a Curie temperature $T_C$ of 15.5 K, as a function of temperature and magnetic field, provided a variety of results which, in particular, revealed an intimate link between the electronic properties and the bulk magnetization. Most informative in this respect were results from measurements of the polar Kerr rotation $\theta_K$, which is directly related to variations of the electronic excitation spectrum upon spontaneous and field induced magnetic order. Kerr rotation spectroscopy also yields information on electronic transitions involving both localized electronic orbits as well as itinerant charge carriers. The $\theta_K$ spectra of EuB$_6$ exhibit two particular features. The first, situated in the infrared spectral range at approximately 0.3 eV, is a resonance which increases in magnitude and shifts to higher energies with either decreasing temperature or increasing magnetic field. At all temperatures and fields this resonance coincides with the plasma edge feature of the optical reflectivity $R(\omega)$ (Refs. 1 and 2) and is giant at low temperatures and high magnetic fields, i.e., of about 11 degree of rotation at 2 K and 7 T (Ref. 3). The second feature in $\theta_K(\omega)$ is a signal at 1 eV, which also grows with decreasing temperature or increasing magnetic field, but does not shift in energy.

The resonance at 0.3 eV is caused by the response of the itinerant charge carriers to magnetism, while the Kerr resonance at 1 eV is associated with the MO response of the 4f-5d interband transitions. A phenomenological analysis, based on the classical Lorentz-Drude dispersion theory adapted to the Kerr rotation phenomenon, revealed that the giant Kerr rotation pinned to the reflectivity plasma edge is indeed the consequence of an interplay between the response of the localized 4f-states and the itinerant charge carriers, a suggestion previously put forward by Feil and Haas. The polar Kerr rotation at 1 eV correlates directly with the bulk magnetization $M(H)$, confirming that the magnetic moments are essentially due to the localized f-electrons of the Eu$^{2+}$ ions. It is thus clear that Kerr spectroscopy allows for distinguishing between the roles of itinerant and localized electrons with respect to magnetic properties of a metal.

In order to confirm this scenario and its interpretation, we have repeated this type of experiments on material of the $Eu_{1-x}Ca_xB_6$ series and on YbB$_6$. It is well established that replacing Eu in EuB$_6$ by isoelectronic Ca changes both the magnetic and the transport properties of these hexaborides. Not surprisingly, the Curie temperature $T_C$ decreases significantly with increasing Ca content. YbB$_6$ was chosen because it provides the possibility to probe the MO response of an essentially non magnetic hexaboride with divalent cations. In YbB$_6$, Yb enters in its divalent configuration with all the 14 f orbits occupied and thus without ionic magnetic moment.

In our measurements of the Kerr rotation in the polar configuration, i.e., with the light beam oriented parallel to the magnetic field, linearly polarized light, a superposition of left (LCP) and right (RCP) circularly polarized components of equal magnitude, is reflected from the sample at a given temperature between 2 and 10 K and in external magnetic fields from 0 to 10 T (Refs. 3 and 11). The polarization plane of the reflected light, with respect to that of the incident light, is rotated by a field and temperature dependent angle $\theta_K$. This is a consequence of the differing absorptions for the two circular polarizations. The Kerr angle is given by

$$\theta_K = -\frac{1}{2}(\Delta_+ - \Delta_-) = -Im\left(\frac{\hat{n}_+ - \hat{n}_-}{\hat{n}_+\hat{n}_- - 1}\right)$$

(1)
where $\Delta_{\pm}$ are the phases of the complex reflectance $r_{\pm}(\omega) = \rho_{\pm} e^{i\Delta_{\pm}}$ and $\tilde{n}_{\pm} = n_{\pm} - ik_{\pm}$ are the complex refraction indices for LCP (−) and RCP (+) light, respectively.\textsuperscript{3,4,11} The relation between $\theta_K$ and the refraction indices in eq. (1) is strictly valid only for Kerr rotations not exceeding 15 degrees, which is the case here.\textsuperscript{11}

The single-crystalline samples of Eu$_{1-x}$Ca$_x$B$_6$, as well as YbB$_6$ were prepared by solution growth from flux, using the necessary high-purity elements as starting materials. All samples were characterized by X-ray diffraction, dc-transport and thermodynamic experiments probing thermal properties.\textsuperscript{8} From magneto-transport data on Ca-doped EuB$_6$, the presence of Al inclusions in the samples can be excluded.\textsuperscript{8} We have investigated two specimens with 20% and 40% Ca-content and Curie temperatures $T_C$ of 5.3 $K$ and 4.5 $K$, respectively.\textsuperscript{8,9}

Figure 1 summarizes $\theta_K(\omega)$ at 2 $K$ and 7 $T$ for 20% and 40% Ca-doping. For the purpose of comparison we also display $\theta_K(\omega)$ for EuB$_6$ (Ref. 3), measured under the same experimental conditions. The most obvious observation is that by Ca-doping the huge resonance at about 0.3 $eV$ is essentially wiped out, whereas the feature at about 1 $eV$ retains its shape and amplitude and shifts only slightly in energy. This feature in $\theta_K(\omega)$ is absent in the results obtained for YbB$_6$, as may be seen in the inset of Fig. 1. Figure 2 emphasizes the field and temperature dependence of the 1 $eV$ resonance of the Eu-based compounds with $x=0.2$ and 0.4. The general trend is the same for both samples and bears similarities with the results for $x=0$. The 1 $eV$ feature increases in magnitude with decreasing temperature and increasing field, but does not exhibit a shift of the resonance frequency. The increase in magnitude tends to saturate at low temperatures and high magnetic fields.

Magneto-optical reflectivity data, to be presented and discussed elsewhere, indicate that Ca-doping induces an enhancement of the scattering rate for itinerant charge carriers, in accordance with dc transport results.\textsuperscript{6} This in turn leads to a so-called overdamped behaviour of the reflectivity, particularly at high temperatures and low fields. The sluggish onset of the $R(\omega)$ plasma edge behaviour in Ca-doped EuB$_6$ is apparent in Fig. 3, where $R(\omega)$ at 10 $K$ and 7 $T$ for the binary compound is also shown for comparison. The $R(\omega)$ plasma edge in Eu$_{1-x}$Ca$_x$B$_6$ for $x \neq 0$ extends over some range in $\omega$ around 0.3 $eV$ and the considerable blue-shift of the edge, as observed for EuB$_6$ (Refs. 1 and 2) with decreasing temperature or increasing field, is absent.\textsuperscript{13}

As we have recently demonstrated,\textsuperscript{4,11} the extended Lorentz-Drude model is quite successful, in reproducing the MO features of EuB$_6$. Therefore, for the analysis of the data presented here we have adopted the same approach as that employed in Ref. 4. It turns out that for the samples of the Eu$_{1-x}$Ca$_x$B$_6$ series with $x \neq 0$, two Drude components are necessary to account for the low frequency electrodynamic response.\textsuperscript{13} Only one of the Drude terms is really relevant for determining the dc $(\omega \to 0)$ transport properties and it also dominates the temperature and magnetic field dependences of $\sigma_1(\omega)$ at low frequencies. The second Drude-type contribution ac-
EuB$_6$, at 0.8 and 1.3 eV for 40% Ca-doping, and at 3.4 and $\sim$ 11 eV for both Ca-contents. The energy splitting of the h.o.’s around 1 eV is consistent with the experimentally verified splitting of the f-states.

Since above 2.5 eV no temperature and magnetic field dependence of the optical properties were detected, the parameters of the Lorentz h.o.’s at 3.4 and 11 eV were established at 10 K, in order to fit the measured reflectivity $R(\omega)$ as well as all the optical functions up to 12 eV. For the fits to the Kerr rotation data in the relevant energy interval (0.23-4 eV), these latter parameters were then fixed for all temperatures and magnetic fields. The parameters of the Drude components and of the two h.o.’s necessary for describing the 1 eV feature in $\theta_K$ were, however, allowed to vary as a function of temperature and field$^{15}$. The overall features of the MO properties and particularly their temperature and magnetic field dependences can be reproduced quite accurately. Figure 3 exemplifies the fit quality for the sample with 20% Ca-content at 10 K and two selected fields. Similarly good fits (not shown here) were obtained$^{11}$ for Eu$_{1-x}$Ca$_x$B$_6$ with $x=0.4$. Figure 3 also demonstrates that the same set of parameters$^{15}$ can equally well account for the fits of $\theta_K(\omega)$, the reflectivity $R(\omega)$ and the real part $\sigma_1(\omega)$ of the optical conductivity. Another information that may be obtained from Fig. 3 is the correspondence between the 1 eV signal in $\theta_K(\omega)$ and the related interband absorption, manifested by the broad feature in $R(\omega)$ and a shoulder in $\sigma_1(\omega)$ at the same energy.

The reduced number of 4f states upon Ca-doping is manifest in the zero-field $\sigma_1(\omega)$ curves of Eu$_{1-x}$Ca$_x$B$_6$ in the spectral range around 1 eV. The shoulder in $\sigma_1(\omega)$ at 1 eV (inset of Fig. 3c), associated with the 4f-5d interband transitions$^{3,4,17}$, decreases with Ca-doping. The total absence of the 1 eV feature in $\theta_K(\omega)$ for the non-magnetic reference material YbB$_6$ (inset of Fig. 1) confirms the validity of the interpretation of this feature. A Kerr rotation is only encountered, if the absorption coefficients for LCP and RCP light are unequal. This situation is not expected for YbB$_6$ with fully occupied 4f electron shells of the Yb$^{2+}$ ions.

The calculated $\theta_K(\omega)$ (Fig. 3a) exhibits no sharp resonance in the spectral range of the reflectivity plasma edge (Fig. 3b) and no blue-shift of the same, neither within nor below the experimentally accessible range for the measurements of $\theta_K(\omega)$ (thin dotted line in Fig. 3a). Thus it agrees with the experimentally observed $\theta_K(\omega)$. In EuB$_6$ the sharp onset of the $R(\omega)$ plasma edge and its blue shift$^2$ were unambiguously correlated with the $\theta_K$ resonance at about 0.3 eV (Ref. 3). This interpretation was supported by the Lorentz-Drude fit even for energies extending to below the experimental limit of the Kerr spectrometer$^{3,4}$. The overdamped behaviour of the $R(\omega)$ plasma edge of Ca-doped EuB$_6$ (Fig. 3b) suppresses the

![FIG. 3: Comparison of the measured and calculated (Lorentz-Drude model) Kerr rotation, reflectivity and real part $\sigma_1(\omega)$ of the optical conductivity at 10 K and selected fields for Eu$_{0.8}$Ca$_{0.2}$B$_6$. The low energy limit of the Kerr spectrometer (i.e., 0.23 eV) is marked with a vertical thin dotted line in panel (a). For the purpose of clarity, the panels (b) and (c) show only the 7 T data, since the field dependence is negligible in this spectral range, as is confirmed by the corresponding calculated curves. For comparison, the sharp onset of the $R(\omega)$ plasma edge of EuB$_6$ at 10 K and 7 T is also shown in panel (b). The inset in panel (c) shows $\sigma_1(\omega)$ for Eu$_{1-x}$Ca$_x$B$_6$ in the spectral range around 1 eV. For all panels the same symbols for $x=0.2$ as those shown in panel (b) apply.](image-url)
necessary resonance conditions, as postulated by Feil and Haas\(^6\), for a giant response in \(\theta_K(\omega)\). Based on magneto-transport data it was suggested that in \(Ca\)-doped \(EuB_6\) the formation of magnetic domain walls is favored by the disorder on the cation sublattice\(^8\). This is probably reflected in the large damping (scattering rate) of the itinerant charge carriers. Upon \(Ca\)-doping the sharp onset of the \(R(\omega)\) plasma edge and the related Kerr resonance, as observed for \(EuB_6\) (Refs. 2,3,4), are broadened and wiped out, respectively.

The relevant quantity of the Kerr spectroscopy, which is selectively sensitive to spin-polarized states, is the relative distribution of spectral weight, i.e., the unequal absorption coefficients, involved in the optical transitions for RCP and LCP light. It is parameterized by the so-called weight factors \(f^\pm\) (Refs. 4 and 11) which shape the dispersion-like curve of \(\theta_K(\omega)\) at 1 eV. We have introduced the quantity

\[
\sigma_f^z = |(f^+ - f^-)/(f^+ + f^-)| = |f^+ - 1|, \tag{2}
\]

with the condition that \(f^+ + f^- = 2\), because of the sum rule constraints. For \(EuB_6\) we have demonstrated\(^4,11\) that \(\sigma_f^z\) is phenomenologically related to the magnetization \(M(H)\), thus acting as the MO counterpart of the moment polarization. Inspection of the \(f^\pm\) factors\(^5\) for the \(h.o.\)’s describing the 1 eV feature and a similar analysis in terms of \(\sigma_f^z\) for the \(Ca\)-doped \(EuB_6\) (Fig. 4) confirm that the 1 eV feature in \(\theta_K(\omega)\) is again associated with the MO response of the 4f electron states which are also responsible for the magnetization of the system\(^7\). In fact, \(\sigma_f^z(H)\) mimics the trend of the magnetization (Fig. 4), just as for \(EuB_6\) (Ref. 4). As for \(EuB_6\), the strongly scattered itinerant charge carriers in the \(Ca\)-doped compounds are hardly polarized (i.e., \(f^\pm \approx 1\)), indicating once more their minor role in shaping the magnetization. The flat \(\theta_K\) spectrum in \(YbB_6\) at 1 eV implies \(f^\pm = 1\) and consequently \(\sigma_f^z = 0\), known as a so-called diamagnetic situation with respect to the Kerr rotation\(^11\).

Taken together, our MO data on \(Eu_{1-x}Ca_xB_6\) and \(YbB_6\) indicate one successful way for achieving large Kerr rotations. Essential are electronic excitations out of incompletely filled 4f electron orbitals. Their effect may significantly be enhanced by excitations of itinerant electrons but it is required that the corresponding scattering relaxation rate is low enough to ascertain a steep slope at the onset of the Drude plasma edge in the reflectivity, thus confirming an earlier suggestion of Feil and Haas\(^5\).

Acknowledgments

The authors wish to thank A. Perucchi, G.A. Wigger and R. Monnier for fruitful discussions. This work has been supported by the Swiss National Foundation for the Scientific Research.

---

1. L. Degiorgi, E. Felder, H.R. Ott, J.L. Sarrao and Z. Fisk, Phys. Rev. Lett. 79, 5134 (1997).
2. S. Broderick, B. Ruzicka, L. Degiorgi, H.R. Ott, J.L. Sarrao and Z. Fisk, Phys. Rev. B 65, 121102(R) (2002).
3. S. Broderick, L. Degiorgi, H.R. Ott, J.L. Sarrao and Z. Fisk, Eur. Phys. J. B 27, 3 (2002).
4. S. Broderick, L. Degiorgi, H.R. Ott, J.L. Sarrao and Z. Fisk, Eur. Phys. J. B 33, 47 (2003).
5. H. Feil and C. Haas, Phys. Rev. Lett. 58, 65 (1987).
6. J. Schoenes and W. Reim, Phys. Rev. Lett. 60, 1985 (1988) and H. Feil and C. Haas, Phys. Rev. Lett. 60, 1985 (1988).
7. W. Henggeler, H.R. Ott, D.P. Young and Z. Fisk, Solid State Commun. 108, 929 (1998).
8. G.A. Wigger, Ch. Wüti, H.R. Ott, A.D. Bianchi and Z. Fisk, Phys. Rev. B 66, 212410 (2002).
9. S. Paschen, D. Pushin, M. Schlatter, P. Vonlanthen, H.R. Ott, D.P. Young and Z. Fisk, Phys. Rev. B 61, 4174 (2000).
10. J.M. Tarascon, J. Etoirneau, P. Dordor, P. Hagenmuller, M. Kasaya and J.M.D. Coey, J. Appl. Phys. 51, 574 (1980).
11. S. Broderick, PhD. Thesis Nr. 14969, ETH Zurich.
12. At 7 T we observe a weak feature in \(\theta_K(\omega)\) of \(YbB_6\) at about 0.5 eV, which does not follow a consistent behaviour either in temperature or in field. We believe that it is due to scattering of the data.
13. A. Perucchi, G. Caimi, H.R. Ott, L. Degiorgi, A.D. Bianchi and Z. Fisk, cond-mat/0307469.
14. F. Wooten, in "Optical Properties of Solids", (Academic Press, New York, 1972), and M. Dressel and G. Grün, in...
"Electrodynamics of Solids", (Cambridge University Press, 2002).

Tables with the full set of fit parameters valid for the data obtained at 2 and 10 K and all chosen magnetic fields are available at the link: http://www.solidphys.ethz.ch/spectro/suppinfo/EuCaB6-Kerr.pdf.

J.K. Lang, Y. Baer and P.A. Cox, J. Phys. F: Metal Phys. 11, 121 (1981) and R. Monnier, private communication.

S. Kimura, T. Nanba, M. Tomikawa, S. Kunii and T. Kasuya, Phys. Rev. B 46, 12196 (1992).

The magnetization has been measured up to 5.5 T (Ref. 8). By assuming a Brillouin function for the overall behaviour of $M(H,T)$, a simple fit of the measured magnetization data leads to the values of $M$ up to 10 T at different temperatures.