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Giant magneto-optical response of ferromagnetic EuB_6

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Abstract. EuB_6, a ferromagnet with a Curie temperature of $T_C \sim 15$ K, exhibits very large magnetoresistive effects at temperatures both above and below T_C. We present our magneto-optical investigations, including measurements of the reflectivity and Kerr rotation, covering the spectral range from the infrared up to the ultraviolet, as a function of temperature between 1.5 and 20 K and in external magnetic fields from 0 to 10 T. The Kerr rotation at high fields and low temperatures is enormous. The spectra reflect aspects of the large magnetoresistance and are shown to discriminate between the spectroscopic response of localized and itinerant electronic states in the ferromagnetic state.

PACS. 78.20.Ls Magnetooptical effects – 75.50.Cc Other ferromagnetic metals and alloys

The origin of ferromagnetism and the large variety of its manifestations are central and, in many regards, still controversial issues in solid state physics [1–5]. In this context, various recent experimental investigations have revealed new and unexpected aspects of ferromagnetism in hexaborides with divalent cations M^{2+} [6]. For instance, EuB_6 is an $S = 7/2$ $4f$-electron local-moment ferromagnet with a Curie temperature T_C of 15.5 K, whereas La-doped CaB_6 is a weak ferromagnet with rather small ordered moments of 0.1 μ_B/La or less, but with Curie temperatures in the range between 600 and 1000 K [6,7].

In this paper we focus our attention on EuB_6. According to specific heat data [8,9], the onset of ferromagnetism occurs *via* two-phase transitions and the magnitude of the ordered moments grows only slowly with decreasing temperature [10]. Parallel to a substantial reduction of the electrical resistivity by two orders of magnitude, a significant blue shift of the plasma frequency is observed in the optical reflectivity as the temperature is reduced to below T_C [8]. The unusually drastic response of the conduction electron system to ferromagnetic order is also reflected in large magnetoresistive effects in the range of T_C and below [11]. In view of these phenomena, EuB_6 is obviously a model system for studying the ferromagnetic state, as well as the interplay between localized moments and conduction electrons and their polarization. According to the magnitude and the temperature dependence of the electrical resistivity, EuB_6 is close to a metal-insulator transition and therefore particularly well suited to investigate the role of itinerant electrons in ferromagnetism in general.

We have chosen to measure the magneto-optical (MO) Kerr effect, a suitable experimental tool for investigating the dependence of the electronic excitation spectrum on the degree of magnetic order. This spectroscopy yields information about the electronic transitions involving localized magnetic moments as well as the response associated with the itinerant charge carriers, especially in relation to the ferromagnetic phase transition. First, we found a giant Kerr rotation occurring in the IR frequency range of the free-electron plasma edge of the reflectivity. It increases in magnitude and shifts to higher frequencies with either decreasing temperature or increasing magnetic field, which indicates that the spin polarized itinerant charge carriers are involved. In addition, a Kerr rotation signal at 1 eV follows the magnetic field and temperature dependence of the magnetic susceptibility and thus is associated with the f-electron response. This offers the possibility of discriminating between itinerant and localized optical responses and indeed opens new perspectives in studying ferromagnetism in metals.

The MO-Kerr measurement is an ellipsometric type of spectroscopy, which measures the characteristics of the anisotropy induced by the sample magnetization. Here, we consider a polar geometry where the light beam is parallel to the external magnetic field. Linearly polarized light, a superposition of left and right circularly polarized components of equal magnitude, is reflected from EuB_6 at a given temperature between 1.5 and 20 K and in an external field from 0 to 10 T. The polarization plane of the reflected light is rotated by a field and temperature dependent angle with respect to that of the incident light. This Kerr rotation is due to the differing absorptions for the two circular polarizations and is expressed by the azimuthal polarization rotation θ_K and the polarization ellipticity ε_K. Here, we present our measurements of θ_K, defined as:

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

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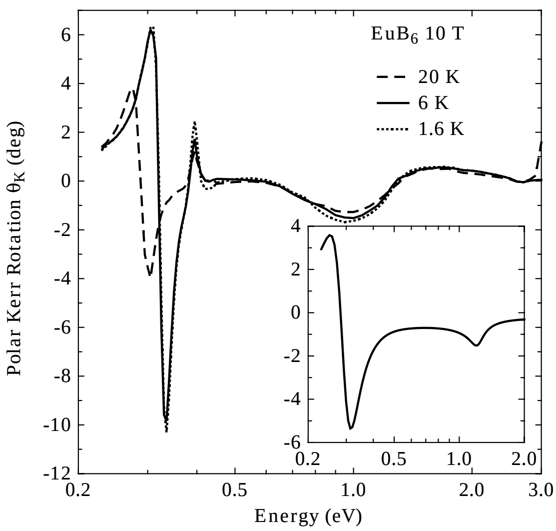

Fig. 1. Kerr rotation spectrum between 0.2 and 3 eV at 10 T and selected temperatures (1.6, 6 and 20 K). (Inset) A corresponding spectrum calculated on the basis of a Drude-Lorentz approach [13,20] and using the following, realistic parameters: optical dielectric constant $\varepsilon_\infty = 1.17$, Drude plasma frequency $\omega_p = 0.75$ eV, Drude scattering rate $\Gamma = 3$ meV, magnetic field $B = 10$ T, Lorentz resonance frequency $\omega_0 = 0.5$ eV, Lorentz oscillator strength $\omega_{p0} = 1$ eV and damping $\Gamma_0 = 0.25$ eV. Obviously, the feature at 0.4 eV was not considered in this simple calculation. Note the logarithmic energy scale.

where $\Delta_\pm$ are the phases in reflectivity and $\tilde{n}_\pm$ are the complex refraction indices for left and right circularly polarized light, respectively [12,13].

Figure 1 gives a flavor of the overall features characterizing the polar Kerr rotation spectra. Displayed are the 10 T data for three selected temperatures at 1.6, 6 and 20 K. Two distinct components may be recognized. First the rather strong and sharp resonance around 0.3 eV, slightly shifting towards higher energies with decreasing temperatures, and a broader signal at about 1 eV. The additional feature at 0.4 eV is not observed in fields of less than 10 T and its amplitude increases with decreasing temperatures. This latter feature will not be further addressed in the discussion. While its sudden appearance at 10 T is rather puzzling, we just conjecture here that it might be a possible MO-manifestation of spin-orbit splitting associated with the sample response at 1 eV.

The field and temperature dependences of the features at 0.3 and 1 eV are displayed in more detail in Figures 2 and 3. In Figure 2, the spectral range around the Kerr rotation resonance at approximately 0.3 eV is emphasized and Figures 2a–c show the field dependence of the resonance at three different temperatures. At 20 K we note a remarkable blue shift and an increasing intensity, *i.e.*, a growing Kerr rotation with increasing magnetic field. The field dependent blue shift is only moderate at 6 K and essentially absent at 1.6 K. At such low temperatures only the intensity of the Kerr rotation increases with increasing magnetic field. The maximum rotation of about $-11°$ at 10 T and 1.6 K, places EuB$_6$ among the compounds with the very largest Kerr rotations ever observed [13].

Another relevant aspect related to the temperature and field dependences of the 0.3 eV resonance is demonstrated in Figures 2d–f, showing the onset of the reflectivity plasma edge as a function of field for the three selected temperatures [14]. We note the similarity between the temperature [8] and the magnetic field dependences of the reflectivity plasma edge. Either with decreasing temperature in zero field [8] or with increasing field at constant temperature (Fig. 2d–f) [14], the plasma edge exhibits an unusual and remarkable blue shift [15]. The field-induced blue shift is considerable at $T > T_C$ but is progressively reduced at the lower temperatures of 6 and 1.6 K. The temperature dependent blue shift of the zero field plasma frequency has been accounted for by an enhancement of the concentration of itinerant charge carriers and a concomitant reduction of their effective mass below T_C [16]. This would suggest a scenario for the ferromagnetic phase transition, where the quasiparticles are progressively undressed with decreasing temperature [5] - a scenario for which the conventional Stoner approach [17] is not adequate. Therefore, a scenario based only on the exchange splitting of the conduction band [15] is not enough, in order to fully explain the data. Considering the present data set it seems natural to conclude that the 0.3 eV Kerr rotation is tied to the reflectivity plasma edge. The enormous MO response at 0.3 eV, as well as the related blue shift of the plasma edge give a direct spectroscopic view on the dynamical response of the free charge carriers in the ferromagnetic state and therefore correlate with features of the (large) magnetoresistance [18].

Figure 3 highlights the second feature in the polar Kerr rotation at energies around 1 eV. This resonance does not shift in energy with either decreasing temperature or increasing field, but its intensity does change with field. At 20 K the polar Kerr rotation progressively increases with increasing fields and saturates above 7 T. In the case of 6 and 1.6 K the intensity saturation occurs already at fields as low as 1 T. The feature at about 1 eV is ascribed to an electronic interband transition, involving the localized f-electron states of the Eu^{2+} ions. The optical conductivity exhibits a variety of features between 1 and 10 eV [8,19]. In particular, the absorption at 1 eV was ascribed to an exciton-type transition between localized $4f$ and more extended $5d(e_g)$ states [19]. Above T_C, *i.e.*, at 20 K, the spin polarization of the localized $4f$ states is induced by the magnetic field, while below T_C the spontaneous ferromagnetic order fully spin-polarizes these states. This scenario accounts for the progressive growth of the 1 eV resonance at 20 K and the saturation of its intensity as a function of magnetic field at 6 and 1.6 K. At 20 K a high magnetic field is indeed required for inducing a Zeeman splitting of the localized states, larger than the thermal energy scale set by the temperature. Consequently, the spin polarization and the corresponding Kerr rotation grow gradually. Below T_C relatively low fields are sufficient to fully spin-polarize the states and thus the Kerr rotation does not grow significantly with increasing fields.

Using a rather simple and phenomenological Lorentz-Drude approach, it can be shown that the two MO

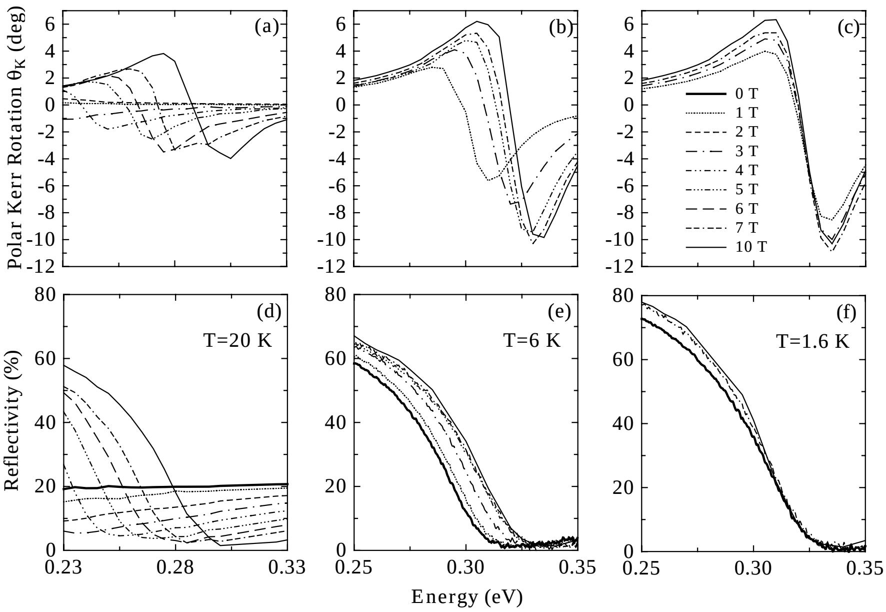

Fig. 2. (Upper row) Kerr rotation spectra emphasizing the resonance around 0.3 eV at (a) 20, (b) 6 and (c) 1.6 K. (Lower row) Corresponding reflectivity spectra in the energy range of the plasma edge onset [8] as a function of magnetic field (0–10 T) and at (d) 20, (e) 6 and (f) 1.6 K. The same line symbols, as shown in part (c) of the figure, are valid at each temperature for both reflectivity and Kerr rotation. Note that for clarity reasons the x-axis scales of part (a) and (d) (*i.e.*, T = 20 K) are not the same as at 1.6 and 6 K. The zero tesla data are obviously shown for the reflectivity only.

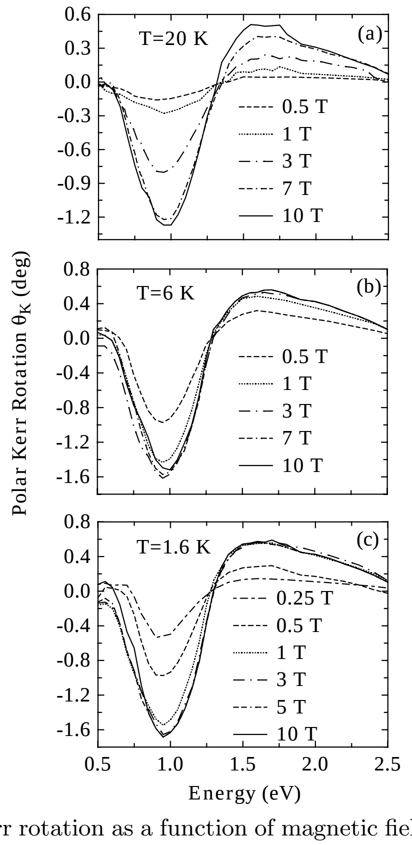

Fig. 3. Kerr rotation as a function of magnetic field for (a) 20, (b) 6 and (c) 1.6 K in the spectral range around the resonance at 1 eV.

responses at 0.3 and 1 eV are interrelated. For this purpose we rely on previous work [20] where the Lorentz-Drude model, based on the classical dispersion theory [21], has been generalized in order to calculate the full off-diagonal conductivity tensor. The interplay between a localized electron interband transition, described with a Lorentz oscillator, and the free-electron response in the form of a Drude term, leads to a strong resonance in the polar Kerr rotation at energies coinciding with the reflectivity plasma edge [13,20], a situation first suggested by Feil and Haas [22] but not entirely recognized or correctly interpreted in previous work [12,23]. In the inset of Figure 1 we show the result of such a calculation. The parameters entering this simple two component calculation (*i.e.*, for the features at 0.3 and 1 eV only), chosen such that the calculated curve approximately reflects the measured signal at 20 K and 10 T, turn out to be quite realistic (see figure caption). More details regarding such calculations and a discussion of their significance will be presented elsewhere [24]. Here, we just mention that there is a difference in the spectral weight of the transitions associated with left and right circularly polarized light [20]. This means that the Kerr rotation data can be reproduced only by considering a so-called paramagnetic type absorption [12,20].

In order to further justify our assignment of the two principal Kerr rotation features to excitations involving itinerant (0.3 eV) and localized (1 eV) electronic states we compare, at different temperatures, the field dependence of the two Kerr rotation peak to peak amplitudes with the field-induced bulk magnetization, as shown for a selection of data in Figure 4. The Kerr amplitudes at 0.3 and 1 eV are normalized by the respective values measured at 1.6 K and 10 T, which are assumed to be the saturation values for both features. In this respect, unity on the right-hand y-axis of Figure 4 coincides with the saturation

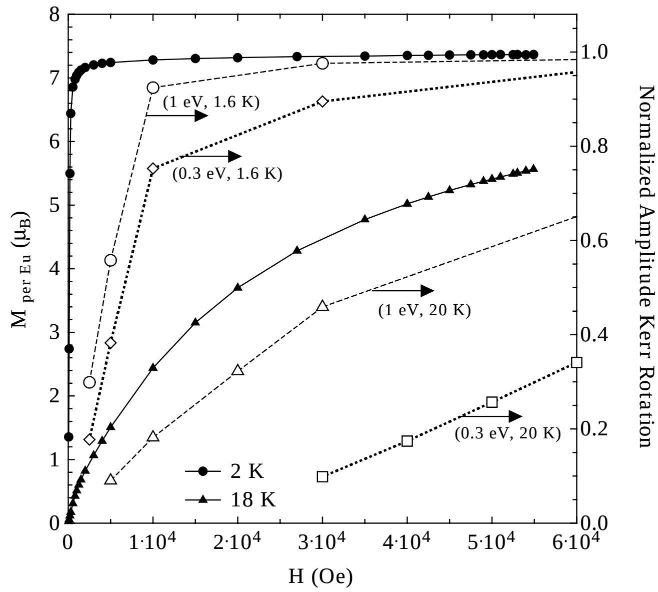

Fig. 4. Magnetization per Eu ion at selected temperatures compared with the normalized peak-to-peak amplitude of the Kerr rotation resonances at 0.3 and 1 eV (see text). The normalized Kerr rotations at 20 K and 1.6 K are compared with the 18 K and 2 K magnetization, respectively. The lines are to guide the eye.

magnetization of $\sim 7.3\ \mu_B$ in EuB_6 [10]. The magnetization $M(H)$ is plotted as the magnetic moment per Eu ion. Although we only show data taken at 2 and 20 K, we note that the general trend of the scaled Kerr rotation at 1 eV is the same as that of $M(H)$ at all temperatures. At the same time it may be seen that the field-induced growth of the renormalized Kerr amplitude at 0.3 eV is much more gradual than that of the 1 eV signal. This observation is consistent with our interpretation of associating the 1 eV signal with transitions between localized $4f$ and more extended $5d$ electron states. These are certainly directly influenced by the bulk magnetization, which is dominated by the polarization of the local $4f$-electron moments [10]. On the other hand, we relate the Kerr feature at 0.3 eV, being tied to the reflectivity plasma edge, to the itinerant charge carriers, which are spin polarized in the ferromagnetic state. The more gradual increase of the corresponding Kerr amplitude with respect to the magnetization curves, particularly at higher temperatures, thus reflects the only indirect influence of the internal field due to local moments on the polarization of the conduction electrons.

In conclusion, the magneto-optical response of EuB_6 exhibits distinct features at different energies, which are both strongly dependent on temperature and external magnetic fields. It reflects microscopic aspects of the very large magnetoresistance effects that are related to the ferromagnetic phase transition and the shift of this transition with external magnetic fields, not reported in this clear form before. We can correlate the MO-response of the itinerant and localized state to the magnetization. Thus, we have gained a spectroscopic access to the magnetic state of the system, and we can identify the direct consequences of ferromagnetism on relevant quantities, such as, the plasma frequency. In a subsequent publication [24] we plan to use these data for extracting, besides the spin polarization of localized and itinerant electrons, the relative variation of the quasiparticles' effective masses in the majority and minority spin bands, triggered by the ferromagnetic phase transition.

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14. The temperature [8] and field dependence of the reflectivity was measured from the far infrared up to the visible spectral range. The complete set of data will be presented and discussed in full detail elsewhere.
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