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Magneto-optical investigations of URhAl intermetallic compound

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Abstract

The uranium ternary intermetallic compound URhAl was investigated by means of magneto-optical Kerr effect. Comparative studies of UTX complexes support the conclusion that the magneto-optical spectrum originates mostly from uranium electronic states, the contribution of other metal states being less important. The most prominent peak revealed in the infrared region of the magneto-optical spectra was attributed to the transitions from the ground 5f states to higher d states of uranium. © 1998 Published by Elsevier Science S.A.

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1. Introduction

Some uranium compounds exhibit a large magneto-optical Kerr effect (MOKE) [1] which is an order of magnitude higher than that observed in current magnetic metals. The search for strong Kerr rotators is motivated first of all by the magneto-optical recording. Although the uranium systems are not very suitable for practical applications because of the low ordering temperature of most of these compounds, the magneto-optics provides valuable information on the electronic structure of studied materials. The investigation of these compounds gives insight into the general rules relating the optical properties to the underlying electron structure.

The aim of our research is the detailed investigation of the electronic structure of UTX compounds (T, transition metal; X, other s or p metal) using optical and magneto-optical methods. In this paper we focus on the magneto-optical properties of the URhAl system and discuss the experimental results based on comparison with other compounds.

The ternary intermetallic compound URhAl crystallizes in the hexagonal ZrNiAl-type structure, space group $P\bar{6}m2$. The atoms are arranged in layers normal to the *c*-axis: a U–Rh layer alternates with a Rh–Al layer [2]. This compound is ferromagnetic below $T_c = 27$ K. The magnetic properties are highly anisotropic which, in part, arise from the hybridization of the uranium 5f and the rhodium 4d electrons. The magnetic moments of the uranium, $\mu_U = 0.94\ \mu_B$, inside one layer are rigidly ferromagnetically coupled due to a strong intralayer exchange and magnetization is oriented along the easy hexagonal *c*-axis [3]. The uranium orbital magnetic moment is higher than the spin moment by a factor of 1.8. Surprisingly, the neutron-scattering experiments indicated a substantial moment also on the Rh atoms situated inside the U–Rh planes, $\mu_{Rh} = 0.28\ \mu_B$ [3]. This implies that the 4d electrons in Rh are highly polarizable due to the hybridization between 5f(U)–4d(Rh) electrons. In contrast, the moments on the Rh atoms inside the Rh–Al plane are almost zero within the experimental error.

2. Experiment

The magneto-optical (MO) measurements were performed on a single crystal of URhAl prepared by a modified Czochralski method in a tri-arc furnace. The Kerr effect was measured in the polar configuration parallel to the hexagonal *c*-axis at near normal incidence (at an angle of 7°). Thin plates for MO measurements were mechanically polished and the magnetic field was applied normal to the sample surface along the easy *c*-axis. Both polar Kerr rotation (PKR) and ellipticity (PKE) were obtained in the spectral range 0.7–5.1 eV (0.24–1.8 μm) and at temperatures from 6 to 35 K. The rotating polarizer/phase plate method provided Kerr angles with resolution better than 0.005°. The measurements were performed in a superconducting solenoid Spectromag II from Oxford

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Instruments. To eliminate the Faraday rotation in windows of the cryostat, the Kerr effect of a sample was compared with that of a reference gold mirror. To minimize the effect of a surface oxide layer, all measurements were made immediately after the polishing, even though exposure to air and, thus, some surface oxidation could not be avoided. Nevertheless, the polished crystal had a mirror surface even after a year exposure to air, with no coloured oxide layer visible with the naked eye.

3. Results and discussion

The sample of URhAl exhibits narrow magneto-optic hysteresis loops with fairly low coercivity (Fig. 1). The coercive field reaches 0.028 T at 6 K and decreases towards 0.007 T at 20 K. Due to the shape anisotropy, the magnetic saturation of MOKE is achieved in non-zero magnetic fields and we can estimate the magnetization from the value of saturation field B_s. At T=6 K we have $B_s \approx 0.15$ T, which gives, for saturated magnetization, $\mu_s = 0.9 \pm 0.06$ μ_B/f.u. The error comes mainly from the uncertainty in the value of the demagnetizing factor. This value of μ_s is in excellent agreement with that obtained from magnetization measurement, $\mu_s = 0.94$ μ_B/f.u. at 4.2 K [4], and also with that calculated from neutron-scattering experiments [3]. This result supports the presumption that the magnetic ordering is not destroyed by the mechanical polishing in the crystal surface layer. The good quality of the sample surface is necessary for reliable measurements of the spectral dependencies of the MOKE.

The spectra of the magneto-optical polar Kerr effect are displayed in Fig. 2. They were measured at T=10 K and B=1 T, i.e. well below the temperature of the magnetic ordering and above the magnetic saturation. For comparison, the spectra of the isostructural UNiGa compound [5] are also displayed in the same plot. Both the rotation and ellipticity are negative in the investigated spectral range with several broad maxima and minima. Measuring the PKR and PKE, i.e. both the real and imaginary components of the complex Kerr effect, enables us to calculate the PKR spectrum from the PKE spectrum and vice versa using the modified Kramers–Kronig relations for the complex MOKE [6]. This procedure gives two useful outcomes: first, the calculations provide the MOKE spectra, even outside the experimental region and, second, it enables to verify the reality of the measured structure in the Kerr spectra. The calculated PKR and PKE spectra (lines) are also displayed in Fig. 2. The most spectacular effect of these calculations is a pair of peaks predicted in the near infrared range in the rotation and ellipticity curves.

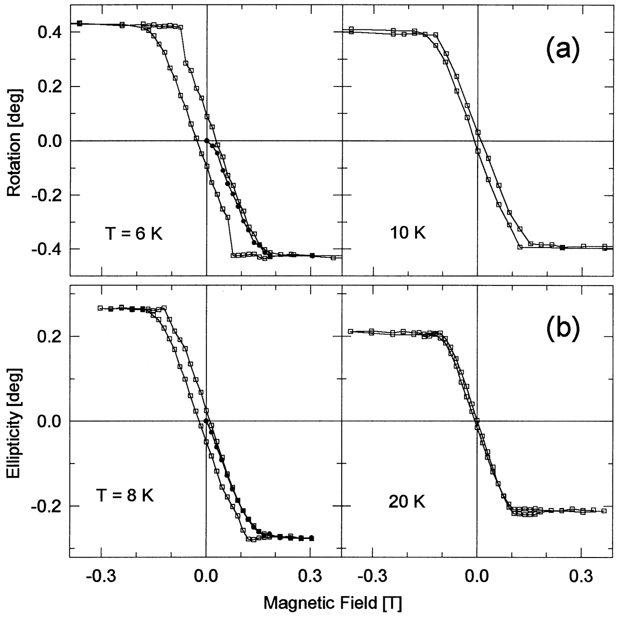

Fig. 1. Polar Kerr rotation (a) and ellipticity (b) hysteresis loops of the URhAl at 2.75 eV (450 nm).

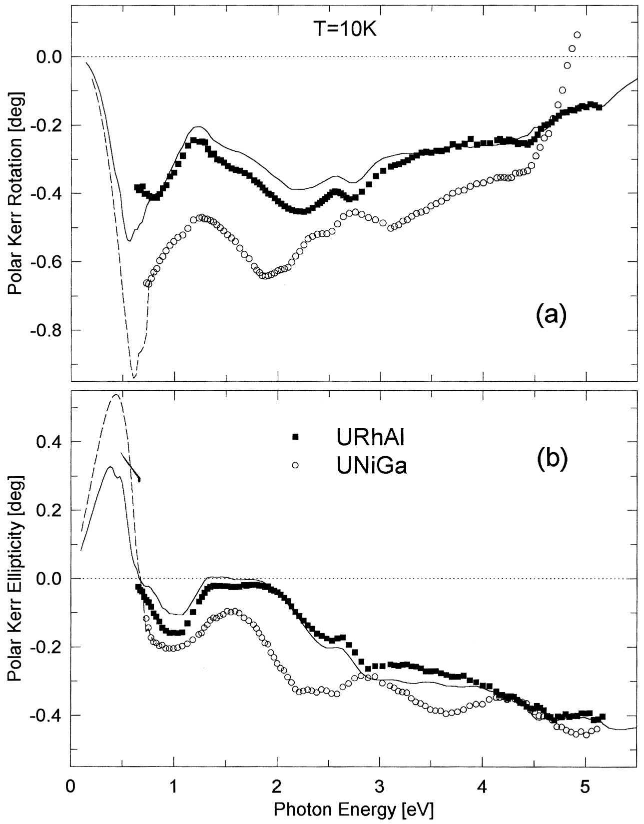

Fig. 2. Polar Kerr rotation (a) and ellipticity (b) of the polished URhAl single crystal. The MOKE spectra of the polished UNiGa are displayed for comparison. The spectra were measured at T=10 K and above the magnetic saturation, at B=1 T for URhAl and at B=6 T for UNiGa. Symbols, experiment; solid and dashed lines, recalculated spectra using the Kramers–Kronig transformation.

A remarkable structure observed on the MOKE curves indicates several interband transitions in the spectral range under investigation. Strong transitions extend to low photon energies and overlap the intraband contribution even in the near infrared region. However, the most prominent peaks were revealed by the Kramers–Kronig analysis in the infrared region near 0.5 eV. It should be mentioned that the shape, position, and magnitudes of these peaks depend on the precision of measuring the Kerr rotation and ellipticity values close to the low energy experimental limit. Nevertheless such peaks can be expected in MO spectra in view of the following facts: (i) the Fermi energy E_F is situated close to the top of the 5f band in the UTX compounds, (ii) this 5f band is highly populated with high joint density of states at E_F (specific heat coefficient is $\gamma=60$ mJ K^{-2} mol^{-1}), and (iii) the photoemission experiments indeed reveal the 5f electron states in the vicinity of the Fermi level in most of uranium compounds [7]. Taking into account these arguments, we can tentatively attribute the infrared pair of peaks to transitions involving partly occupied ground 5f states and higher 6d states of uranium, 5f→6d. This conclusion should be, however, supported by experimental measurements of MOKE at lower energies. It is worth noting that the dominating peak in the near infrared region is a common feature of most of uranium intermetallics.

The magneto-optical properties of URhAl strongly depend on the band structure and the correlation with calculations of the density of states (DOS) is necessary for more reliable interpretation of the spectra at higher energies (visible and ultraviolet ranges). The bands above 1 eV should obviously involve also the 4d states of rhodium which was reported to bear some magnetic moment [3] and also sp metal states.

It is interesting to compare the MOKE spectra of URhAl and UNiGa (Fig. 2). The absolute value of the Kerr angles is higher in UNiGa in all the spectral range, however, a striking resemblance of both spectra is apparent in spite of different chemical composition of these two compounds. Especially, the structure at low energies up to 2 eV is almost identical. Thus we can infer that the main contribution to the MO effects originates mostly from the uranium electronic states, the contributions from the transition metals T or from the X-metals are less significant in the discussed UTX compounds. Moreover the correspondence of low-energy bands in spectra suggests that the DOS distribution near E_F is similar in both compounds, and is determined predominantly by the 5f states of uranium. These findings are interesting in view of the different magnetic properties of the transition metals in both compounds. While those Rh atoms within the U–Rh layers bear a magnetic moment which is oriented parallel to the total moment of uranium [3], the Ni atoms are commonly reported as nonmagnetic in UNiGa[1]. Besides, the strength of the spin-orbit interaction for the Rh atoms is high compared to the 3d transition metals [8] and, because the MOKE value increases approximately linearly with the spin-orbit interaction parameter [8,9], we could expect also a certain contribution to the MOKE spectra coming from the Rh electronic states. In fact, huge MO rotations were observed in some Rh-doped magnetic garnets [10] demonstrating a crucial role of the Rh electronic states in the MO effects which makes these atoms the most efficient MO dopants among the d metals. Nevertheless, this is not the case of the discussed UTX compounds. Taking into consideration these arguments, the Rh substitution does not lead to substantial changes of the MO spectra in uranium intermetallics in the spectral range under investigation, at least in UTX systems, and the spectra are strongly influenced first of all by the electronic states near E_F, i.e. by the uranium states. It is worth noting that the role of the X-metal in the MO spectra is rather indirect—it mainly adjusts the interatomic distances and changes the overlap of the electron wave functions of the neighboring atoms, which is though essential for the f–d hybridization effects.

In this respect it is not surprising that the shape of the optical spectra at low energies is similar. Indeed, a wide maximum usually dominates in the near infrared region, with regard to the number of actinide and rare-earth metal compounds which are characterized by high density of states near E_F. Thus, the identification of the lowest situated strong peaks in the optical spectra as 5f→6d transitions is thought to be reasonable.

On the other hand, the intraband contribution originating from various scattering processes of free carriers (coming from transitions assisted by scattering at lattice imperfections and phonons), which is expected to dominate in the infrared range, cannot be reliably distinguished in the MOKE spectra. The preliminary reflectivity measurements manifest that the free carrier contribution is significant only at very low photon energies, so that we deduce that this contribution is rather weak in the experimental region of our MO measurements.

4. Conclusions

The magneto-optical investigations of the URhAl intermetallic compound support the hypothesis that the structure in the magneto-optical spectrum originates mostly from uranium electronic states. The contribution of Rh and Al states is less significant. The most prominent peak revealed using Kramers–Kronig transformation in the infrared region was attributed to the transitions from the ground 5f states to the higher 6d states of uranium. However, these peaks should be supposed as speculative for the time being, unless they are supported by experiment. The structure in the MOKE spectra strongly depends on the electronic band structure of the investigated compound and the correlation with ab initio relativistic calculations of the electronic structure is necessary for more precise interpretation of the optical properties.

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[1] Recent polarized neutron experiments on UNiGa suggest that some small magnetic moment is also detected on Ni atoms situated within the Ni–Ga layers (M. Olsovec, private communication).

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