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Magneto-optic properties of uranium-based compounds

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Uranium-based compounds often show interesting magnetic properties and large polar Kerr rotations. Because of this, the wavelength and temperature dependencies of the polar Kerr rotation in several uranium-based compounds, including UMn_2Ge_2, UFe_2, and UGa_2, have been investigated. The Mn moments order ferromagnetically in UMn_2Ge_2 below 380 K, and while the U moments do not order ferromagnetically until the temperature is below 150 K. The measurements presented here show that the size of the Kerr rotation increases from 0.05° at room temperature to 0.15° when the U moments are ordered. The wavelength dependence of the Kerr rotation between 500 and 1000 nm is rather weak. UGa_2 orders ferromagnetically at 130 K, and a Kerr rotation of 0.2° at 85 K and 632.8 nm can be measured, with the rotation increasing to 0.4° at 1000 nm. UFe_2 orders ferromagnetically at 180 K, and the Kerr rotation (85 K, 632.8 nm) is 0.17°. These magnetic and optical properties are discussed in terms of the electronic states of uranium.

I. INTRODUCTION

Several U-based compounds, most notably the chalcogenides[1] and the pnictides,[2] have very large Kerr rotations below their magnetic ordering temperatures. The large Kerr rotations presumably arise because U is a heavy atom with a large spin-orbit interaction and because of its propensity to form *f* bands near the Fermi surface, which in turn can lead to allowed optical transitions in the visible or near-infrared spectral region. Both effects are necessary if the compound is to exhibit sizable magneto-optic properties. Because of this, we have investigated the magnetooptic properties of three U-based compounds at temperatures below their ordering temperatures.

II. EXPERIMENT

Bulk polycrystalline samples of UMn_2Ge_2, UFe_2, and UGa_2 were prepared by arc-melting in an inert gas atmosphere. Some samples were further processed by wrapping them in Ta foil and annealing at 800 °C for one week. X-ray-diffraction measurements showed that the samples were single phase and of the desired structures. Kerr rotation measurements were made on mechanically polished samples using apparatus previously described.[3] Kerr rotation measurements were made for wavelengths between 500 and 1100 nm, and for temperatures between 80 and 400 K. Measurements at low or high temperatures were limited to magnetic fields less than 8 kOe.

III. RESULTS AND DISCUSSION

The magnetic properties of UMn_2Ge_2 have been quite thoroughly investigated. UMn_2Ge_2 has the $ThCr_2Si_2$ tetragonal structure, with planes of U atoms lying perpendicular to the crystalline *c* axis. Magnetization measurements by Dirkmaat *et al.*[4] show that the Mn moments order ferromagnetically at 380 K, with very little magnetic anisotropy. The U moments, in contrast, have their magnetic easy axis along the crystalline *c* axis. As the temperature is lowered, the U-Mn interaction tends to align the Mn moments along the *c* axis. The ferromagnetic ordering of the U sublattice is apparently quite sluggish, occurring gradually for temperatures below about 150 K. van Engelen, de Mooij, and Buschow[5] have reported a Kerr rotation of 0.05° at room temperature, and by comparison with $LaMn_2Ge_2$, propose that most of the rotation is due to the U moments. Figure 1(a) shows a Kerr hysteresis loop (632.8 nm) for UMn_2Ge_2 at 85 K. The sample appears to be nearly saturated by 8 kOe, and the Kerr rotation (at 8 kOe) is about 0.15°. Some variation in the size of the Kerr rotation was observed for different samples, apparently due to texturing in the samples. The temperature dependence of the Kerr rotation is shown in Fig. 2. The Kerr rotation increases from zero as the sample temperature is lowered from 400 K, and it reaches a plateau between room temperature and 200 K. The room-temperature rotation is 0.05°, in agreement with the results of van Engelen and co-workers.[5] As the temperature is reduced from 200 K, the Kerr rotation again increases, which we believe is due to ordering of the U moments. This result differs from the conclusions of van Engelen and co-workers, who proposed that most of the room-temperature Kerr rotation is due to

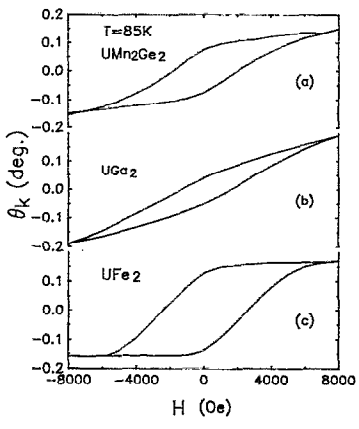

FIG. 1. Kerr rotation hysteresis loops of several uranium compounds at 85 K using light of wavelength 632.8 nm: (a) UMn_2Ge_2, (b) UFe_2, and (c) UGa_2. For each compound, the sign of the Kerr rotation is negative.

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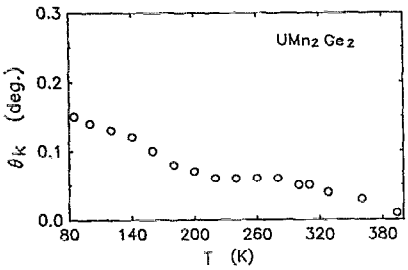

FIG. 2. Temperature dependence of the Kerr rotation of UMn_2Ge_2 using light of wavelength 632.8 nm and an applied field of 8 kOe. The increase in Kerr rotation below 200 K is apparently due to ordering of the U moments.

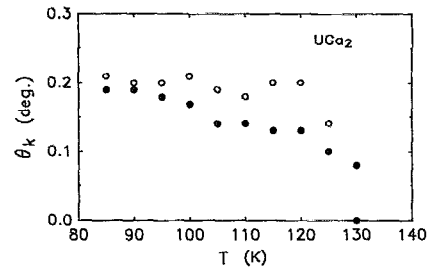

FIG. 4. Temperature dependence of the Kerr rotation of UGa_2. The two different symbols correspond to measurements on two different samples. The differences between these two samples are attributed to differences in sample texture and composition. Note that the samples were not saturated in these measurements.

the U moments. The results of Fig. 2 offer clear evidence that the U moments do not order until the temperature is less than 200 K.

UFe_2 forms in the cubic (C-15 type) Laves phase. The transition temperature seems to be strongly dependent upon stoichiometry, with stoichiometric samples having a Curie temperature above 190 K, while Fe-deficient samples have Curie temperatures as low as 150 K.[6,7] The magnetic easy axis is along $\langle 111 \rangle$.[6] Neutron-diffraction measurements on polycrystalline and powder samples show that most of the moment resides on the Fe atoms, with only 10% of the moment on the U atoms.[6,8] A Kerr rotation hysteresis loop for UFe_2 at 85 K is shown in Fig. 1(b). The sample appears to be saturated, with a rotation of 0.17° at 8 kOe. The temperature dependence of the Kerr rotation is shown in Fig. 3. Our sample appears to have a transition temperature near 180 K, suggesting that it is reasonably stoichometric.

UGa_2 has the hexagonal AlB_2 structure at room temperature.[9] Below the Curie temperature of 130 K, the structure becomes orthorhombic, apparently because of a large magnetostrictive interaction.[10] The moment on the U atoms is 2.7 μ_B at 4.2 K. Fig. 1(c) shows the Kerr rotation hysteresis loop (632.8 nm) of UGa_2 at 85 K. This appears to be a minor loop. Above the Curie temperature, the Kerr rotation depends linearly on the field, indicating a highly-paramagnetic behavior. Figure 4 shows the temperature dependence of the Kerr rotation (8 kOe, 632.8 nm) for two different samples cut from the same arc-melted button. The two samples have quite different Kerr rotations, and slightly different transition temperatures, suggesting slight compositional and texture differences. A third sample showed a Kerr rotation of 0.4° at 85 K, but annealing reduced this to 0.2°.

Figure 5 shows the wavelength dependencies of the Kerr rotation for all three samples. The Kerr rotations for these samples vary by about a factor of 2 over the visible to near-infrared spectral region, with little structure. The sizes of the Kerr rotations are relatively modest, being an order of magnitude smaller than those observed in other U-based compounds. Figure 6 shows measured values of the real and imaginary parts of the dielectric constant of each of the samples. These measurments were obtained using an ellisometry system that has been described elsewhere.[11] As can be seen, ϵ_1 and ϵ_2 vary smoothly over the visible spectrum for all three samples. The absence of structure is consistent with our wavelength-dependent Kerr rotation measurements.

In summary, we have measured the Kerr rotations in three U-based compounds, UMn_2Ge_2, UFe_2, and UGa_2. Several U compounds have shown large Kerr rotations, apparently due to the presence of U-induced visible and near-infrared interband transitions and a large spin-orbit interaction because of the heavy U atoms. The compounds investigated here only show modest Kerr rotations. The interpretation of the magnetic and magneto-optic properties of these compounds would be aided by a knowledge of

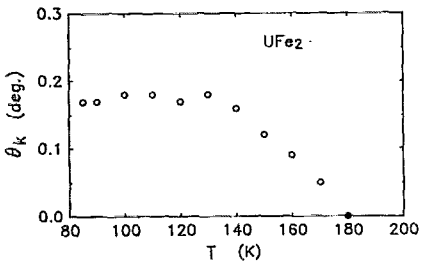

FIG. 3. Temperature dependence of the Kerr rotation of UFe_2 using light of wavelength 632.8 nm and an applied field of 8 kOe.

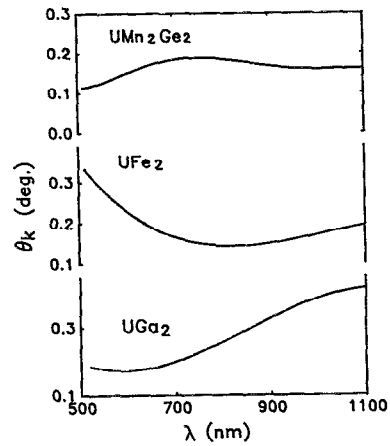

FIG. 5. Wavelength dependencies of the Kerr rotation for (a) UMn_2Ge_2, (b) UFe_2, and (c) UGa_2.

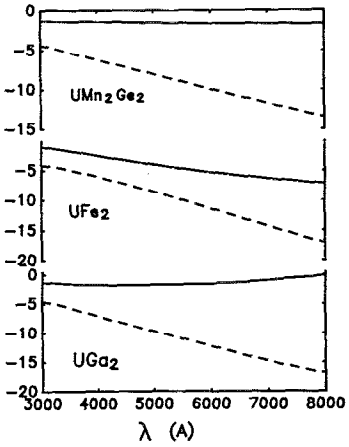

FIG. 6. Real (ϵ_1) and imaginary (ϵ_2) parts of the dielectric constants of UMn_2Ge_2, UFe_2, and UGa_2 at room temperature. The solid lines correspond to ϵ_1, while the dashed lines correspond to ϵ_2.

their electronic structures. In particular, it would be of interest to know the location of the U $5f$ levels and the presence of unoccupied d states which would permit f-d transitions in the spectral region of interest. Thus, photoemission measurements, which are planned for the future, and self-consistent spin-polarized band calculations may help to achieve a more detailed understanding of the magnetooptic properties of these compounds.

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