Magneto-Optical Evidence of Exchange-Induced Moment in Uranium at Room Temperature Observed in Co/U-As Multilayers

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The magneto-optical Kerr rotation was measured in Co/U-As multilayers and compared with calculations of the Kerr response of such multilayered structures. From the thickness dependence of the Kerr rotation it is concluded that the U-As layers have an exchange-induced magnetic moment at room temperature reaching 20% of the low-temperature value.

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Since the discovery of large magneto-optical (MO) effects in U compounds by Schoenes and Reim [1–3], a key question has been whether there exist materials containing U that are magnetic at room temperature with an appreciable magnetic moment on the U. In this event, the U can be expected to contribute to the MO response at room temperature. Reim and Schoenes pointed out [3] that, so far, there are only a few materials in which a contribution of U at room temperature to the MO spectra was supposed. Examples are $UMnGe_2$ and $UMnSi_2$, where Van Engelen, de Mooij, and Buschow [4] assume a U moment at room temperature based on a comparison with the corresponding La compounds. Neutron diffraction experiments [5], on the other hand, indicate the absence of a moment on U in these materials. This means that the origin of the Kerr rotation at room temperature could be of another origin than that assumed by Van Engelen.

In analogy to Co/Pt multilayer films, where a polarization is induced in the Pt through exchange coupling to Co layers [6], we studied the possibility of exchange coupling in sputtered multilayer films of Co and U-As layers. Based on our work on ferromagnetic amorphous U-As thin films [7,8] with Curie temperature $T_C \leq 140$ K, we investigated Co/U-As multilayers with a fixed [U]/[As] ratio of 1.5 with Co and U-As film thickness ranging from 2 to 80 Å. First results on the crystallographic and magnetic properties of films with Co layer thickness $t_{Co}=10$ Å were presented before [9]. For $t_{Co} \geq 10$ Å the films are ferromagnetic at 300 K. Multilayer films with thinner Co layers do not show magnetic ordering at room temperature because of interdiffusion of U and As into the Co [9]. By preparing films with $t_{Co}=20$ Å and with varying U-As layer thickness $t_{UAs}=2$–80 Å we have obtained MO evidence of an induced magnetic moment in U at room temperature through exchange coupling to Co layers.

The films are prepared by magnetron sputtering from the elements at sputtering rates of 1.3–1.9 Å/s onto glass substrates at ambient temperature as described in detail in Ref. [7]. The structures are deposited under computer control. The Co layers are prepared by holding the glass substrates over the source and opening the shutter for a precalibrated time; the U-As alloy layers are prepared by sequentially depositing subatomic layers of 1 and 1.5 Å of U and As, respectively, to give a ratio [U]/[As] $\simeq 1.5$. A Co buffer layer of 200 Å is deposited first onto the substrate in order to assure crsytalline growth of the Co. The number of bilayers Co/U-As is varied from 14 to 170 to give a total film thickness of 1400 Å. The topmost layer is always Co in order to prevent corrosion of the underlying U-As layers. Low-angle x-ray diffractometry is used to confirm the periodicity of the structure. Corresponding spectra are shown in Ref. [9]. The total weight of Co, U, and As is determined chemically [7] by dissolving an entire codeposited film or by Rutherford backscattering.

The polar Kerr-effect measurements are carried out from the film side of the samples using a standard lock-in technique [3]. An Oxford optical Dewar with a split-coil superconducting magnet (7 T magnetic field) is used for the low-temperature measurements at 10 K; the room-temperature measurements are made without a Dewar using a water-cooled magnet providing a maximum field of 2.8 T. Both setups are fully computerized. A reference measurement, using an Al or Pt mirror, is subtracted from the sample measurement to account for the Faraday rotation of the Dewar windows or optical elements. The data are usually taken in both positive and negative fields and averaged in order to subtract any birefringence of nonmagnetic origin.

Films with Co and U-As layer thickness ranging from 2 to 80 Å are measured in the photon energy range 0.7–5 eV. For Co layer thickness $t_{Co} < 10$ Å no long-range ordering is found at room temperature. Interdiffusion of U and As into the Co layers is assumed to be responsible for the suppression of magnetic ordering. A comparative study on U-As-Co amorphous alloys [10] corroborates these findings. Films with $t_{Co}=10$ Å are magnetic at 300 K but have only a small Kerr rotation θ_K ($|\theta_K| < 0.15°$) which increases considerably at low temperatures, reaching $-0.5°$ in Co(10 Å)/U-As(30 Å) at 10 K [9].

For $t_{Co}=20$ Å, θ_K is plotted in Fig. 1 for varying U-As layer thickness t_{UAs}. It is evident that the U-As layers

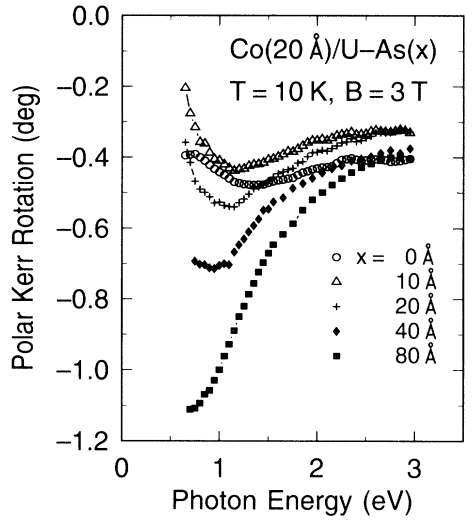

FIG. 1. The polar Kerr rotation of Co(20 Å)/U-As(x) multilayer films at a temperature of 10 K and in a field of 3 T.

develop a strong moment at 10 K. Taking a maximum θ_K of $-1.65°$ for a $U_{61}As_{39}$ composition [8], we calculate for a Co(20 Å)/U-As(80 Å) film a rotation of $-1.3°$ in good agreement with the measured value of $-1.1°$. The slightly smaller measured value is attributed again to the loss of magnetic moment in the U-As layers by interdiffusion. Hence, the U-As layers develop the full magnetic moment at low temperature as compared with the amorphous binary alloys. Polar Kerr rotation loops show some hysteresis, indicating the random anisotropic nature of amorphous U-As layers at 10 K.

Figure 2 shows the Kerr rotation θ_K and ellipticity ε_K at 300 K for some selected multilayer films. The low-temperature spectra of crystalline UAs and of an amorphous $U_{61}As_{39}$ thin film are given in the insets of Fig. 2. For $t_{UAs}=2$ Å, the MO response is smaller than in pure Co as expected from dilution and interdiffusion. At $t_{UAs}=20$ Å, a completely different shape of the spectra is observed. The Co peak at 3.8 eV has disappeared and the peak at 1.3 eV is shifted to lower energies. A comparison with the inset reveals a striking resemblance with the spectra of the binary UAs compounds at low temperatures. Such a change in the spectral dependence of the MO response can be caused by two effects, an optical-constant effect or an active magnetic moment on the U or both. The optical-constant effect originates in the change in relative index of refraction $\tilde{n}=\tilde{n}_{Co}/\tilde{n}_{UAs}$ at the Co/U-As interface as compared to a Co/vacuum interface, where $\tilde{n}=\tilde{n}_{Co}$. This leads to different prefactors $A=n^3-3nk^2-n$ and $B=-k^3+3n^2k-k$, where $\tilde{n}=n-ik$, which connect the off-diagonal conductivity, $\tilde{\sigma}_{xy}=\sigma_{1xy}+i\sigma_{2xy}$, to θ_K and ε_K,

$$\theta_K=\frac{4\pi}{\omega}\frac{B\sigma_{1xy}+A\sigma_{2xy}}{A^2+B^2}, \tag{1}$$

$$\varepsilon_K=\frac{4\pi}{\omega}\frac{A\sigma_{1xy}-B\sigma_{2xy}}{A^2+B^2}. \tag{2}$$

In order to elucidate the role of these two effects, we have calculated the Kerr spectra for various t_{UAs} using a matrix formalism [11,12]. The basic idea is to sum up the contributions of the individual layers taking into account the attenuation and Faraday rotation within the layers. This matrix formalism has been successfully applied in the past to Co/Pd and Co/Au multilayers [13,14]. The input data for the U-As layers, θ_K, ε_K, n, and k, are taken from values of crystalline UAs (Refs. [2] and [15]). The Co data are taken from Ref. [16].

The results of the calculations, assuming no magnetic moment on the U, i.e., a pure optical-constant effect, are

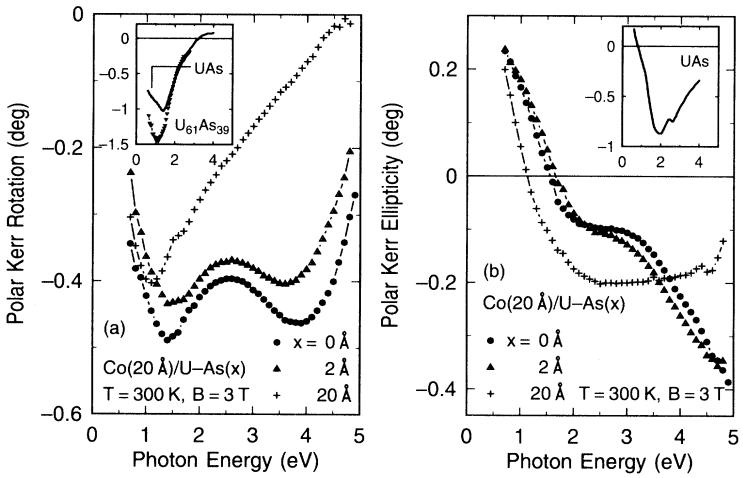

FIG. 2. (a) The polar Kerr rotation and (b) ellipticity of Co(20 Å)/U-As(x) multilayer films at a temperature of 300 K and in a field of 3 T. Inset: The polar Kerr rotation and ellipticity for the crystalline UAs compound (Ref. [2]) and for a $U_{61}As_{39}$ amorphous thin film (Ref. [8]) at a temperature of 20 and 10 K, respectively.

shown in Fig. 3(b), together with the corresponding measurements [Fig. 3(a)]. It becomes evident that the disappearance of the high-energy peak of Co is due to the optical-constant effect alone. The calculations do not account for the shift of the low-energy peak to lower energies as observed in the measurements for increasing t_{UAs}. This is because the data of the crystalline NaCl-structure compound UAs are used in the calculations whereas amorphous U-As thin films exhibit a peak around 1 eV [8]. The absolute value of the low-energy peak, however, is calculated to decrease almost linearly with increasing t_{UAs}, in apparent contrast to the measurements where a leveling off is observed.

Figure 4 emphasizes this discrepancy by plotting the maximum Kerr rotation θ_{Kp} at (a) 10 K and (b) 300 K as a function of U-As-layer thickness t_{UAs}. Figure 4(a) compares the low-temperature values of θ_{Kp} with a calculation, θ_{fm}, taking into account, besides the optical-constant effect, the full magnetic moment on U (solid line). For small $t_{UAs} \leq 10$ Å, the measured absolute values of θ_{Kp} decrease with thickness whereas the calculation increases monotonically over the whole thickness range. This means that about 10 Å of each U-As layer interdiffuses with the Co layers forming at the interface a U-As-Co alloy which has a strongly suppressed magnetic moment [10]. The difference between theory and experiment at large t_{UAs} is due to the fact that the calculations used the data for the crystalline NaCl compound UAs, which has a magnetization of $0.42\mu_B$ per U atom. This is only 60% of the magnetization of amorphous U-As films ($0.7\mu_B$ per U atom) [8]. If the MO data of crystalline UAs is scaled with the magnetization of amorphous U-As films and the effect of a 10-Å layer of U-As-Co alloy at each interface is taken into account as a reduction of the magnetic moment in the U-As layers, a very good agreement is achieved between the measurements at 10 K and the calculation θ_{calc} [dashed line in Fig. 4(a)], where

$$\theta_{calc} = \begin{cases} \theta_0, & t_{UAs} \leq 10\ \text{Å}, \\ \theta_0 + (\theta_{fm} - \theta_0)\dfrac{t_{UAs} - 10\ \text{Å}}{t_{UAs}}\dfrac{0.7}{0.42}, & t_{UAs} > 10\ \text{Å}. \end{cases} \tag{3}$$

θ_0 is the calculated rotation assuming an optical-constant effect only. Note that within our model the U-As-Co alloy is assumed to influence only the magnetic properties of the U-As layers and not the optical properties because interdiffusion creates a gradual change in the optical functions from Co-like to U-As-like. Therefore, the interface is simply washed out, which we neglect in our approach.

At 3000 K, θ_{Kp} is compared with a calculation θ_0 (solid line) taking into account an optical-constant effect only, as plotted in Fig. 4(b). The formation of an U-As-Co alloy at the interface shows up as a rapid decrease in θ_{Kp} for small $t_{UAs} \leq 10$ Å. For $t_{UAs} \geq 20$ Å, however, the measured θ_{Kp} is significantly larger than the optical-

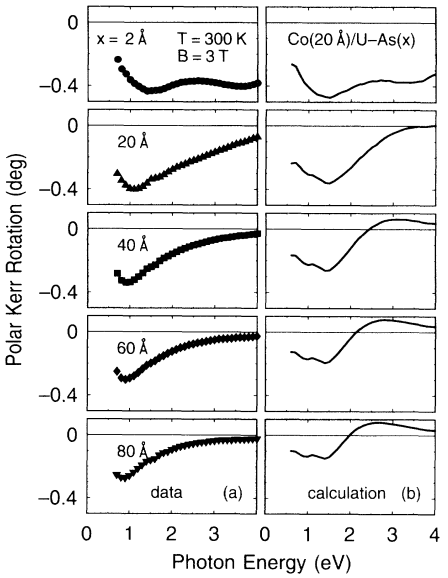

FIG. 3. (a) The polar Kerr rotation of Co(20 Å)/U-As(x) multilayer films at a temperature of 300 K. (b) The calculated Kerr rotation for a multilayer structure with the same parameters as in (a) assuming an optical-constant effect but no MO activity in the U-As layers.

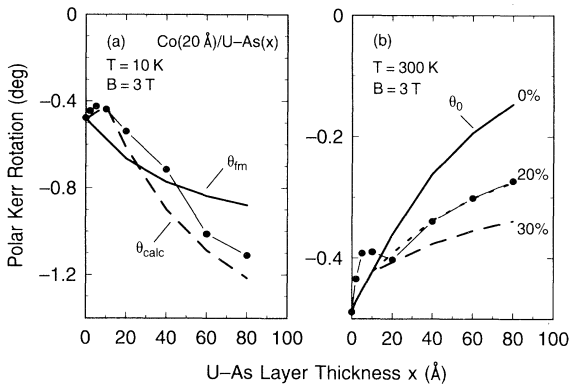

FIG. 4. The maximum polar Kerr rotation of Co(20 Å)/U-As(x) multilayer films at a temperature of (a) 10 K and (b) 300 K as a function of U-As layer thickness. The solid line in (a) is the calculated maximum rotation θ_{fm} taking into account the optical-constant effect and the full magnetic moment in the U-As layers. The dashed line in (a), θ_{calc}, takes also into account the formation of 10 Å of a MO inactive U-As-Co alloy at each Co/U-As interface and scales the magnetic moment of the U-As layers to the higher magnetic moment of the amorphous U-As thin films. The solid line in (b) is the calculated maximum rotation θ_0 assuming only an optical-constant effect. The dotted and dashed lines in (b) are the calculated values assuming an exchange-induced magnetic moment in the U-As layers of, respectively, 20% and 30% of the low-temperature value.

constant effect alone would predict, indicating an exchange-induced moment on the U atom. We can calculate the size of the exchange-induced moment in the U-As layers at room temperature by introducing a partial MO activity of the U-As layers, which corresponds to a partial moment on the U, and by taking into account the effect of 10 Å U-As-Co alloy formation at each interface as discussed above:

$$\theta(x_{\mathrm{im}}) = \begin{cases} \theta_0, & t_{\mathrm{UAs}} \leq 10\ \text{Å}, \\ \theta_0 + (\theta_{\mathrm{fm}} - \theta_0)\dfrac{t_{\mathrm{UAs}} - 10\ \text{Å}}{t_{\mathrm{UAs}}}\dfrac{0.7}{0.42}x_{\mathrm{im}}, & t_{\mathrm{UAs}} > 10\ \text{Å}, \end{cases} \tag{4}$$

where x_{im} is the induced magnetic moment on the U as a fraction of the low-temperature value. The dotted and dashed lines in Fig. 4(b) represent the calculations according to Eq. (4) with, respectively, 20% and 30% induced moment on the U. This clearly shows that the exchange-induced moment in the U-As layers amounts at room temperature to 20% of the low-temperature moment of amorphous U-As thin films. This corresponds to $0.14\mu_B$ per U atom which is only about 5% of the total magnetization of the film. Hence, the induced moment on the U has no big effect on the total magnetization, but due to the strong spin-orbit coupling in U, the influence on the MO response is strong and shows as an enhancement in θ_{Kp}. We found from measurements on amorphous U-As films at 300 K a contribution to the Kerr rotation of less than 0.05° from the paramagnetic moment induced in U by an applied field of 3 T. Therefore, the effect on the Kerr response of such a paramagnetic moment of the U is small.

In contrast to the low-temperature measurements, polar Kerr rotation loops do not show any hysteresis at 300 K. They appear as hard-axis loops. In-plane magnetization measurements confirm an easy axis in the plane and coercivities typical for Co.

In summary, we present MO Kerr spectra of Co/U-As multilayer thin films and compare them with a calculation which takes into account the optical-constant effect and a MO activity of the U-As layers. It is concluded that the disappearance of the Co peak at 3.8 eV is due to the optical-constant effect and that the large room-temperature rotation in the multilayer films with thick U-As layers (> 20 Å) is due to an exchange-induced moment in the U reaching 20% of the low-temperature moment of binary U-As amorphous thin films. Therefore, evidence for an induced moment in U at room temperature through exchange-coupling to Co layers is given in these multilayer films.

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