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Magnetic moment of Au at Au/Co interfaces: A direct experimental determination

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X-ray magnetic circular dichroic measurements at the $L_{3,2}$ edges of Au in Co/Au multilayers are reported. A clear evidence for Au polarization is provided. The induced magnetic moment of Au is very small, about one order of magnitude smaller than in Co/Pt multilayers. The ratio of orbital-to-spin magnetic moment of Au is found to be $\mu_L/\mu_S=0.12$. The ferromagnetic response of Au and its role on the interpretation of the magneto-optic spectra of Co/Au multilayers are discussed with respect to *first principle* calculations.

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The last decade we have encountered an enormous progress in Synchrotron Radiation Facilities. Modern third generation Synchrotron Radiation sources provide photons with tunable energy and with a brilliance of 12 orders of magnitude larger than the conventional rotation anode x-ray tubes do.[1] The development of techniques with element specificity and monolayer sensitivity such as the x-ray magnetic circular dichroism (XMCD)[2,3] and x-ray magnetic scattering[4] allows us, nowadays, to probe directly even the small induced magnetic moments at the interfaces between ferromagnetic and nonferromagnetic layers. For transition metal $3d/5d$ multilayers, recent results have revealed the profile of spin induced polarization of Pt,[5] the relative orientation between the magnetic moments of the ferromagnetic and the nonferromagnetic element,[5–7] as well as the relative orientation between the spin and orbital induced magnetic moment of W, Ir, and Pt.[7] Impressive effects such as an apparent "violation" (breakdown) of the third Hund's rule for W in Fe/W multilayers have been demonstrated.[7]

Although some progress in measuring and understanding the induced magnetic moments of $5d$ transition metals at interfaces has already been achieved, recording clear magnetic signals from normal metals at interfaces remains a challenge. This is attributed to the very small induced magnetic moments expected for normal metals. For example, in atomic form Au has completely filled $5d$ states, that is there is no net magnetic moment. In metallic form, there is a small number of d holes in the $5d$ band of Au due to hybridization effects. Proximity with $3d$ ferromagnets at interfaces should then result in exchange spin polarization of Au and the appearance of a finite induced magnetic moment. Already, early theoretical calculations have predicted a very small spin magnetic moment of about 0.03 μ_B/(atom of Au) at the Fe/Au interfaces.[8] Quite a lot of theoretical works have yield numbers for the spin and orbital magnetic moment of Au at interfaces since then, however, due to the small magnitude of these quantities, the published data exhibit a large scattering.[8–11] Interestingly, hybridization, spin-orbit coupling, and the existence of Au induced magnetic moments have been considered to be responsible for the modification of the magneto-optic spectra of Co or Fe at the higher energy range (above 3 eV) in Co/Au and Fe/Au ultrathin films and multilayers.[10,12] However, from an experimental point of view, up to now there are only indirect qualitative results for magnetic polarization of Au at the (Fe, Co, Ni)/Au interfaces. These results include the variation of the hyperfine field of ^{119}Sn probes embedded in Au and measured by Moessbauer spectroscopy[13] and the appearance of features in spin-resolved photoemission spectra of Co films grown epitaxially on a Au(111) single crystal.[14] In Refs. 13 and 14, neither the magnitude of the total magnetic moment of Au, nor separation into spin and orbital contributions were provided. In the present work we measure directly, by XMCD, the induced magnetic moment of Au in Co/Au multilayers and separate it into spin and obital terms. The Au magnetic moment is found to be almost one order of magnitude smaller than the induced magnetic moments of other $5d$ transition elements in multilayers. The spin and orbital magnetic moments of Au deduced in this work may serve as a reference for *first principle* calculations, while the existence of a finite Au magnetic moment itself justifies the validity of theoretical models for the interpretation of the magneto-optic spectra of $3d/5d$ element-based multilayers.[10,15,16]

Co_n/Au_m multilayers were prepared by electron (e)-beam evaporation under ultrahigh vacuum (UHV) conditions on Si, glass, and polyimide substrates. (The indices denote numbers of atomic planes in the multilayer period.) Details on the UHV chamber and growth conditions may be found elsewhere.[17] In the beginning of each evaporation a 30 nm buffer layer of Au was grown in order to isolate the multilayer from the substrate and to allow for better crystallinity and {111} texturing. The number N of bilayer repetitions for the multilayers was equal to 30. The multilayer growth was always ending at a 5 nm capping layer for chemical protection of the samples against oxidation and corrosion. The samples were structurally characterized by x-ray diffraction (XRD). We plot in Fig. 1 the experimental XRD spectra (solid lines) for a Co_{12}/Au_4multilayer on Si. On the left-hand side of Fig. 1 one may see the experimental XRD spectrum consisting of many small-angle multilayer diffraction peaks indicating the high degree of reproducibility of the

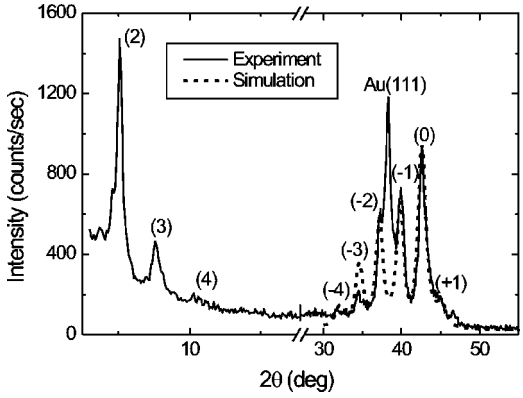

FIG. 1. Experimental (solid) and simulated (dashed line) x-ray diffraction spectra for a Co_{12}/Au_4 multilayer revealing a rectangular-like wavelength modulation profile.

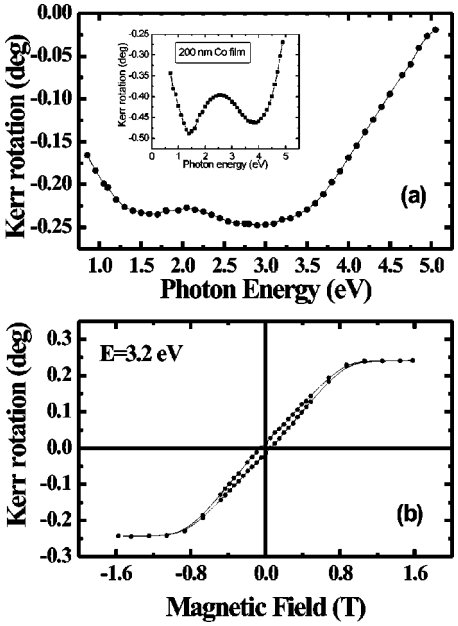

FIG. 2. (a) Polar Kerr rotation spectrum and (b) hysteresis loop for the Co_{12}/Au_4 multilayer at room temperature with the external magnetic field applied perpendicularly to the film plane. The hysteresis loop is mirror inverted in order to be comparable to magnetization hysteresis loops. The inset in (a) shows the corresponding spectrum from a 200 nm Co film measured also at room temperature in the same setup.

multilayer unit cell. On the right-hand side of Fig. 1 the buffer/overlayer Au(111) and an average face-centered-cubic(111) multilayer diffraction (denoted by "0") dominate the spectrum. The observation for the fcc structure of our multilayers is in agreement with recent high resolution electron microscopy studies of Co/Au(111) multilayers.[18] Moreover, the existence of plenty of satellites surrounding the 0 diffraction is a direct indication of the very high quality of the growth of our multilayers. Using the model described in Ref. 19, the diffraction pattern can be modeled with an excellent agreement between experiment (solid) and simulation (dashed line), assuming $n=12.5$, $m=3.9$, bulk-like interlayer spacings $d_{Co}=0.2051$ nm and $d_{Au}=0.2356$ nm, increased (decreased) for Co (Au) by 0.009 nm at the interface. The simulation has revealed that the degree of interdiffusion is negligible, i.e., it is strictly limited at one atomic plane at the interface, as in the case of Ni/Pt multilayers.[4] Our result confirms previous reports on the interface abruptness of Co/Au multilayers prepared by e-beam evaporation or molecular beam epitaxy processes.[20] The smaller intensities of the experimental higher order satellites, as compared to the calculated ones, may be interpreted as a loss of the structural coherence due to the relatively large multilayer thickness of about 105 nm.

The magneto-optic response of the Co/Au multilayers was measured in the energy range of 0.8–5 eV in a magnetic field of 1.64 T. A computer-driven Kerr spectrometer with a resolution of 0.002° was used. The samples were positioned in polar Kerr geometry within the air gap of an electromagnet. As a reference, in order to control the influence of Faraday rotation from the optical components of the setup on our results, an Al mirror was included in the spectra. To cancel out the influence of stress-induced birefringence effects all measurements were taken with both field polarities and subtracted. Polar Kerr hysteresis loops were taken by simply ramping the field at the respective photon energies.

In Fig. 2(a) the polar Kerr rotation spectrum of the Co_{12}/Au_4 multilayer is plotted, whereas the hysteresis curve in Fig. 2(b) shows that our sample is rather hardly magnetized in the direction normal to the film plane (saturation field of about 1 T is needed). The magnitude of Kerr rotation is in fair agreement with previous reports for similar Co/Au multilayers[21] and quite smaller than the one of a Co film measured with the same setup (see inset). The latter effect is attributed to the additivity law in the multilayer[22] and to the existence of a 5 nm capping Au layer on the top of the multilayer. Taking into account the limited penetration depth of about 15 nm at the photon energy range of the experiment, the magnetically active thickness, i.e., the one of Co, that contributes to the Kerr rotation is of about half of the total film thickness. This results in the strong reduction of Kerr rotation as compared to the Co film (inset). Such a dilution effect on the Kerr rotation has also been reported for Co/Au multilayers with relatively thick Au layers.[23] The spectrum of the Co/Au multilayer presents slightly enhanced behavior at higher energies (at about 3 eV) with respect to the lower energies (about 1.5 eV) in comparison to the spectrum of pure Co (inset). This enhanced behavior is in principle similar to the corresponding one of Co/Pt multilayers (especially to the ones with thick Co and thin Pt layers, see Ref. 24) and it is important for technological applications in the magneto-optic recording industry, see for example Refs. 23–25. To interpret this result one has to consider that the enhanced Kerr rotation at higher energies is mainly due to the existence of the large spin-orbit coupling of Au atoms.[15,16] Direct orbital hybridization between Au and Co at the interface produces a spin polarization of Au, while the large spin-orbit coupling of Au is transmitted to Co resulting in the enhanced Kerr rotation maximum at higher energies.[16] For such an interpretation to be valid, it is required for Au to carry some induced magnetic moment and this will be directly demon-

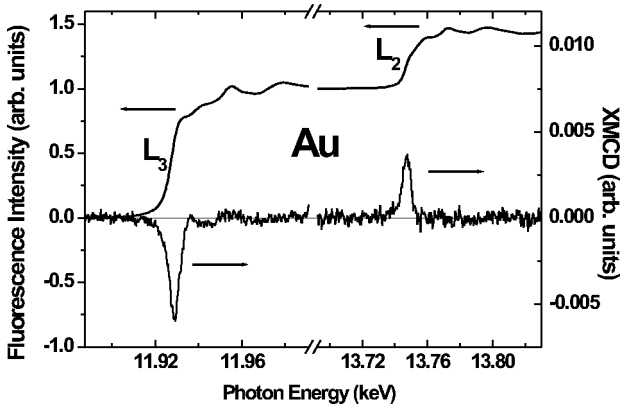

FIG. 3. X-ray absorption (top) and x-ray magnetic circular dichroism (bottom) spectra recorded at the $L_{3,2}$ edges of Au in a Co_{12}/Au_4 multilayer.

strated and quantitatively determined in the following.

The XMCD experiments were performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France) on the ID12 beamline[26] at the $L_{3,2}$ edges of Au using a highly efficient fluorescence yield detection mode in a backscattering geometry. The degree of polarization of the circular light provided by an apple-II type helical undulator HU38 after the monochromator equipped with a pair of Si(111) crystals was nearly 98%. Since the saturation field is 1 T [see Fig. 2(b)], in order to ensure complete magnetic saturation, large magnetic fields of 2 T were applied perpendicular to the film plane. Spectra recorded at 4 and 300 Kshowed no difference due to the very high (bulk-like) Curie temperature of the Co/Au multilayers with relatively thick Co (12 atomic layers of Co in each multilayer period). To exclude any experimental artifacts, the XMCD spectra were recorded either by changing the helicity of the incoming light or by inverting the direction of the external applied magnetic field.

In Fig. 3 the x-ray absorption (XAS) and the XMCD spectra at the $L_{3,2}$ edges of Au in a Co_{12}/Au_4 multilayer are plotted. For the XAS spectra the ratio of the L_3/L_2 was normalized to 2.24/1 according to Ref. 27. The existence of finite XMCD signals shows unambiguously that Au has acquired an induced magnetic moment. The XMCD signals are very small, only of about 7‰ with respect to the XAS, making, previously, the detection of Au polarization at interfaces with ferromagnetic layers, impossible. It is only now that the high photon flux and degree of polarization offered by the third generation Synchrotron Radiation Facilities[26] allow us to record such small XMCD signals with large signal-to-noise ratio. By knowing the direction of the magnetic field and the helicity of the beam we conclude that Au is polarized parallel to the magnetic layer moment, in agreement with *first principle* calculations.[8,10,11]

In order to determine the magnetic moment of Au and separate it into spin and orbital contributions we have applied the sum-rule analysis.[28] The "white line" intensities at the L_3 and L_2 edges were taken from a reference Au_4Mn compound to be equal to 0.374 and 0.183, respectively. For the number of *d* holes for Au we have used the value $n_h = 0.688$.[29] We estimate that this approximation gives a maximum error of about 10%; indeed the number of *d* holes calculated for pure Au is 0.617.[29] Our analysis provides for the total magnetic moment of Au the value $\mu_{tot} = 0.031\ \mu_B$/atom, whereas the ratio of orbital-to-spin magnetic moment is $\mu_L/\mu_S = 0.12$. At this point one has to recall that XMCD probes the average Au magnetic moment in the sample. If one considers that all the Au signal comes strictly from the interface layer then the upper limit for Au polarization at the Co/Au interfaces is 0.062 μ_B/atom. The magnetic moment of Au is of about an order of magnitude smaller than the induced magnetic moments of 5*d* elements in magnetic multilayers, namely W, Ir, and Pt.[30] This is explained by the fact that the 5*d* band of Au is almost full as compared to the aforementioned elements. Guo and Ebert[9] predict a smaller average magnetic moment of $\mu_{tot} = 0.018\ \mu_B$/atom for Au in $Fe_1(001)/Au_5$ due to antiparallel alignment of the Au moments in the first and second from the interface layers. Recent first principle calculations for tetragonal $Fe_1(001)/Au_1$ (Ref. 10) and body-centered-cubic $Fe_5(001)/Au_3$ (Ref. 11) superlattices predict slightly larger values for the induced magnetic moment of Au in the range 0.06–0.08 μ_B/atom. For Fe(001)Au interfaces, previous *first principle* calculations have predicted a spin magnetic moment of 0.03 μ_B/atom for Au.[8] For the ratio of μ_L/μ_S there is quite some disagreement in theoretical works with values ranging between 0.08 (Ref. 11) and 0.6.[9] On the other hand, for Au impurities in bulk Fe theoretical calculations predict much larger induced magnetic moments for Au of about 0.2–0.3 μ_B/atom, while early XMCD experiments in bulk alloy samples have probed $\mu_{tot} \approx 0.1\ \mu_B$/atom.[31] It should be mentioned here that, while calculations for the induced magnetic moment of Au at the Fe/Au interfaces are available, there is no work for Co/Au multilayers. Therefore, we have compared our data to the existing calculations for Fe/Au and we would welcome theoretical works for Co/Au interfaces.

In conclusion, spin, orbital, and total magnetic moments of Au at Au/Co intrfaces were directly probed by x-ray magnetic circular dichroism. The result was compared to previous experimental observations of induced magnetic moments of 5*d* metals in multilayers and may serve as a future reference for *first principle* theoretical calculations. Last but not least, the verification and determination of the existence of a finite Au magnetic moment supports existing models for the interpretation of the magneto-optic spectroscopic behavior of Au-based multilayers at the higher energy range of the spectra.

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