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X-ray scattering and absorption studies of epitaxial strains in Co–Au superlattices

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X-ray scattering and EXAFS are used to probe epitaxial strain in Co–Au superlattices grown by molecular beam epitaxy. We observe a thickness-dependent strain in ultrathin cobalt layers and find that tensile strains near misfit dislocations may be larger than in the more coherent interior of the Co layers. A strong enhancement of the magnetic absorption spectrum is observed in the superlattice samples.

Epitaxial structures based on ultrathin layers of cobalt have been found to exhibit unusually pronounced magnetic anisotropies [1]. Of special interest is the crossover to a perpendicular easy axis of magnetization for cobalt layers thinner than 11 Å in a Au–Co–Au sandwich structure [2] and for Co thicknesses less than ≈ 20 Å in a Co–Au superlattice [3].

Recent studies [4, 5] suggest that *epitaxial strain* plays a key role in the magnetic behavior of these materials. In particular, we have shown [5] that a magneto-elastic contribution to the anisotropy energy can account quantitatively for the thickness dependence of the magnetic anisotropy in Co–Au and Co–Cu superlattices grown by molecular beam epitaxy (MBE).

In this paper we present the results of a combined X-ray diffraction and X-ray absorption study of the epitaxial structure of Co–Au superlattices grown by MBE on GaAs substrates. Here we take advantage of the special characteristics of synchrotron radiation, especially the dispersive EXAFS technique [6] at the LURE DCI storage ring. Subtle differences are observed in the values of strain measured by X-ray scattering and EXAFS. They most probably arise because the former technique emphasizes long-range correlations whereas EXAFS is sensitive mainly to near-neighbor atomic correlations. Thus, since the EXAFS data include contributions from incoherent regions of the heterostructure, EXAFS is a useful probe of the *interface* structure. We expect the latter to be dominated by short-range order as a result of the presence of misfit dislocations [7]. Complementary views provided by EXAFS and diffraction are relevant to understanding the relationship between interface structure and magnetic anisotropy in these systems.

Superlattice samples of Co–Au were grown [3] by MBE on GaAs (110) substrates to a total film thickness of approximately 1500 Å for X-ray diffraction studies and 4000 Å for the EXAFS samples. For the present experiments we selected a series of structures in which the cobalt layer thickness was varied from 5 to 30 Å and the gold spacer layers were of fixed thickness (16 Å). This range spans the crossover to perpendicular anisotropy mentioned above. Of great importance to studies of ultrathin magnetic layers is the abruptness of the interfaces; as immiscible elements, Co and Au are expected to be favorable constituents for superlattice growth. Several techniques, including X-ray scattering [3], high resolution transmission electron microscopy (TEM) [3] and spin-echo NMR [8], provide evidence that the interfaces in Co–Au are indeed very sharp with steps of no more than ± 1 monolayer in height.

X-ray diffraction. The strain parallel to the layers is measured using an X-ray transmission method in which the diffraction vector lies parallel to the plane of the sample. In order to optimize the scattering intensity the GaAs substrate is thinned mechanically to $\approx 100\,\mu m$. Diffraction measurements are performed using a rotating anode X-ray generator (MoKα; $\lambda = 0.71$ Å) with the samples mounted on a Huber four-circle diffractometer.

EXAFS. X-ray absorption measurements were made at the Co K-edge ($E_0 = 7.709$ keV) and at the Au L_{III}-edge ($E_0 = 11.919$ keV). Several pieces ($\approx 5 \times 5\,mm^2$) of the sample were stacked together in order to further increase the absorption. For these measurements the GaAs substrate was completely removed by etching. EXAFS was carried out in the dispersive mode using synchrotron radiation with the linear polarization parallel to the layers. Absorption data were collected over a ≈ 500 eV range simultaneously by means of a curved Si(311) polychromator and a 1-dimensional diode array detector [6].

X-ray dichroism. Magnetic absorption measurements were performed using circularly polarized X-rays emitted at a small angle (0.4 mrad) below the plane of the storage ring. The sample is placed between the poles of an axial electromagnet where the magnetic field is reversed every second so that alternate absorption spectra correspond to a spin alignment parallel (μ^+) and antiparallel (μ^-) to the beam. The difference in the spin-dependent absorption, $\Delta\mu = \mu^+ - \mu^-$, is normalized to $\mu^+ + \mu^-$.

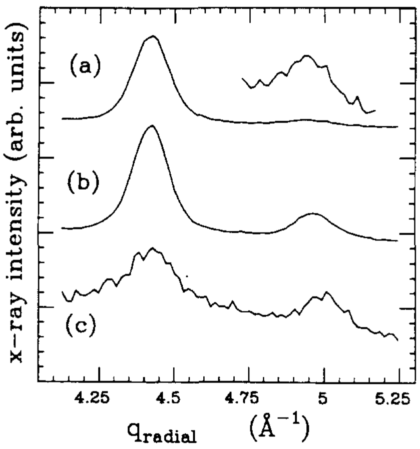

Fig. 1. X-ray scattering measurements performed in transmission geometry with the scattering vector lying in the plane of the superlattice. The total superlattice thickness is 1500 Å and the Au layer thickness is 16 Å in all cases. The Co layer thicknesses are (a) 10 Å, (b) 20 Å and (c) 30 Å. The vertical scale has been adjusted in each case to reveal peak shapes and positions.

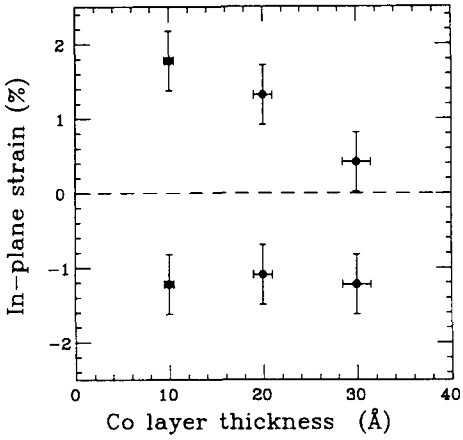

Fig. 2. In-plane epitaxial strains plotted against Co layer thickness, as derived from the peak positions in fig. 1. The Co layer strains are in all cases tensile (upper points) and the Au layer strains are compressive (lower points).

Figs. 1 and 2 summarize the results of in-plane X-ray diffraction scans for various cobalt layer thicknesses. From the positions of the peaks (fig. 1), we can measure the lattice spacing within each layer relative to their bulk values. The Co-layers are found to be under tensile stress, parallel to the layers, whereas Au is compressed relative to the bulk (fig. 2). Note that because of the large ($\approx 14\%$) mismatch between Au and Co, most of the strain is relieved by interfacial misfit dislocations. The epitaxial orientation of the layers, however, is preserved and the resulting in-plane diffraction pattern is a superposition of the separate reciprocal lattices of Au and Co. The in-plane strain in the Co layer is found to be strongly thickness dependent, decreasing as the layer thickness increases. This behavior is well understood in terms of dislocation formation for layers thicker than some critical thickness ($\lesssim 2$ Å in this case).

A recently developed treatment [4] of the magnetic anisotropy shows that the crossover to perpendicular anisotropy can be accounted for quantitatively by magneto-elastic contributions arising from the thickness-dependent strain. A reasonable fit to the observed effective anisotropy is obtained assuming bulk values of the magnetocrystalline and magneto-elastic coefficients of hcp cobalt [5].

The X-ray diffraction measurements described here relate to the *mean* in-plane Co strain averaged primarily over the coherent portion of the cobalt layer structure. It is of interest to compare these values (fig. 2) with those derived from EXAFS measurements which include contributions from incoherent regions of layer structure, i.e., from Co atoms at interface steps and in the vicinity of interface dislocations.

Fig. 3 compares the differential EXAFS spectrum of a bulk Co sample with that of a 30 Å Co/16 Å Au superlattice sample. Marked differences are observed, in particular a shift of the superlattice EXAFS oscillations towards lower energies in the superlattice samples. Since we were not able in this experiment to collect data over a wide enough energy range to make an accurate Fourier inversion of the EXAFS spectrum, we analyzed these energy shifts in terms of a simple relation of the form: $(E - E_0)R^2 =$ constant, where E_0 is the energy of the Co-edge and R is the first-neighbor distance [9]. In this way the relative strain is $\Delta R/R = \frac{1}{2}\Delta E/E$, where

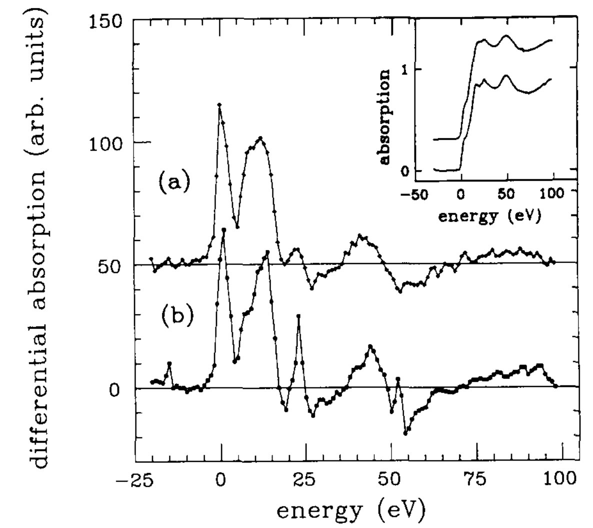

Fig. 3. Differential absorption spectra for (a) a 30 Å Co–16 Å Au superlattice and (b) bulk Co. The corresponding near edge absorption spectra are shown in the inset for the superlattice (upper curve) and for bulk Co (lower curve). A Co layer strain value of 1.5% is derived from the shift in the superlattice differential absorption peak positions to lower energies, relative to the bulk positions.

ΔE is the energy shift. We measured shifts of $\Delta E = 0.33$, 0.66, 1.33 and 1.66 eV, by calculating the median positions of the spectral features at energies $E - E_0 = 11.33$, 22.66, 42.00 and 82.66 eV, respectively. This method returns a value of 1.5% for the tensile strain of Co, significantly larger than the value measured by X-ray diffraction (0.4%) for 30 Å cobalt layers. This comparison suggests that the Co layers neighboring the Co–Au interface are significantly more strained than those in the presumably more coherent interior of the layer. Such a difference could be explained by a slightly less dense packing in the vicinity of misfit dislocations. In this connection there are several different mechanisms that could lead to inhomogeneous strain at the interface. In particular, the results here suggest a residual misfit localized at interface dislocations [10].

The near-edge features of the Co absorption spectrum were also recorded in the presence of a magnetic field (≈ 5 kOe), sufficient to align the magnetization perpendicular to the plane of the film in the 30 ÅCo/16 Å Au superlattice. The idea of this type of experiment is to study the spin density of the final states of the photoelectron. These are related, but not identical to, the ground states of the Kerr effect, of importance in magneto-optic recording applications. It is interesting in this context to note that a large enhancement (5 × bulk) of the specific Faraday rotation has been observed in ultrathin Co–Au films [11]. The difference in absorption, $\Delta\mu$, between the cases where the d-electron spins are parallel or antiparallel to the photoelectron polarization P_e, is [12]:

$$\Delta\mu \sim |M_{1s}(E_x)|^2 P_e \Delta\rho(E),$$

where M_{1s} is the photon transition matrix element and $\Delta\rho = \rho^{\uparrow} - \rho^{\downarrow}$ is the difference in spin density between the majority and minority bands. This effect has been studied by Schütz et al. [13] in several transition metal and rare earth ferromagnets using somewhat different (scanning monochromator) X-ray absorption techniques than those employed here.

In fig. 4 we show the normalized spin-dependent magnetic absorption measured on bulk Co (a foil ≈ 5 μm in thickness) and on a superlattice sample with 30 Å Co layers. In both cases there is a spin polarization extending some 10 eV above the Fermi level. We point out an enhancement in the spin density of states in the superlattice structure. The sign of the signal is that of an empty minority band, as for bulk cobalt. The enhancement may be related to the strain of the cobalt layer and possibly to an increase of the moment borne by cobalt close to the interface. We think that it is unlikely that the enhancement is due to an overlap with states of the neighboring Au layers because similar absorption measurements at the Au L_{III}-edge, gave a null result for $\Delta\mu$ at the level of 1 in 10^{-4}.

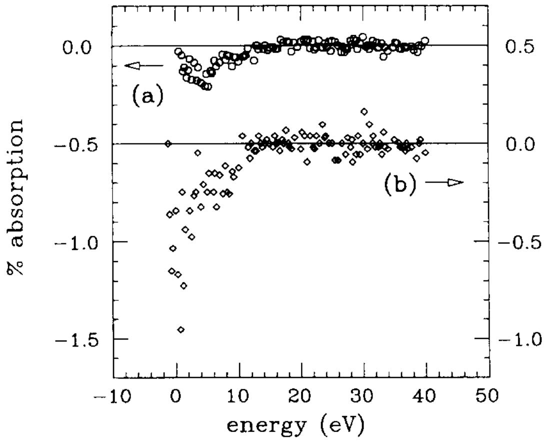

Fig. 4. Spin dependent absorption measured at the Co K edge for (a) bulk Co and (b) 30 Å Co–16 Å Au superlattice. Within 10 eV of the edge the superlattice values are enhanced by a factor of 2–3, relative to those of bulk Co.

In conclusion, we have studied the behavior of epitaxial strain in Co–Au superlattices as a function of cobalt layer thickness. We find that the Co strain parallel to the layers decreases with increasing thickness, a result that is useful in explaining the crossover to perpendicular anisotropy in these materials. Subtle differences in strain as measured by X-ray diffraction and by EXAFS may reflect the different environments of atoms in the interior of the layers and of those neighboring dislocations and steps. An enhancement of the spin density of states of unoccupied levels above the Fermi level is observed in a superlattice sample suggesting that interfacial effects have a pronounced effect on the electronic band structure.

Acknowledgements

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