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Soft-X-ray magnetic circular dichroism: a new technique for probing magnetic properties of magnetic surfaces and ultrathin films

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We demonstrate the feasibility of applying the novel soft-X-ray magnetic circular dichroism (SXMCD) technique to investigate the magnetic properties of magnetic surfaces and ultrathin films. Measurements have been carried out on Ni films of various thickness on a Cu(100) substrate at the Ni $L_{2,3}$ absorption edges. The SXMCD data exhibit strong temperature and thickness dependence, giving film thickness dependent Curie temperatures and suggesting that a single monolayer of Ni on a Cu(100) substrate may be non-magnetic.

1. Introduction

Recent advances in synchrotron radiation instrumentation have made possible the production of high quality circularly polarized light in the soft-X-ray region (100–1500 eV) [1,2]. Soft-X-ray magnetic circular dichroism (SXMCD), the difference in the absorption intensity of magnetic systems using left- and right-circularly polarized soft-X-rays, provides a unique means to probe the element and site specific magnetic moments in a broad range of magnetic systems. It offers new information complementary to that from spin-polarized photoemission spectroscopy, and can resolve ambiguities in absorption studies using unpolarized light.

Magnetic circular dichroism (MCD) measurements using soft-X-rays have several advantages over using photons in other energy regions. The soft-X-ray region gives access to the strong dipole-permitted core 2p to valence 3d excitations of transition-metal and core 3d to valence 4f of rare-earth magnetic materials. These 3d and 4f states are responsible for most of the magnetic properties. In the hard-X-ray region [3] these 3d and 4f states can only be reached, respectively, through the weak quadrupole transitions from the 1s core level of transition-metals and 2p core level of rare-earths. The broad natural linewidth of these deep 1s and 2p core levels also trends to smear out the fine structure in absorption and MCD spectra. On the other hand, in the VUV region [4] the MCD can be measured through the strong dipole-permitted core 3p to valence 3d excitations of transition-metal and core 4d to valence 4f excitations of rare-earths, however, the unresolvable spin-orbit splitting of the $3p_{1/2}$ and $3p_{3/2}$, and of the $4d_{3/2}$ and $4d_{5/2}$ levels make the data analysis a difficult task. In comparison to the well-established magneto-optic Kerr effect technique [5], MCD in the visible light region, the SXMCD process involves only local excitations, and therefore gives element and site specifications and a relatively easier data interpretation than that of the MOKE, where detailed knowledge of the delocalized valence (occupied) and conduction (unoccupied) states together with the momentum dependent optical transition matrix

elements are required. The situation for MOKE is even more complicated for systems where electron correlation effects play an important role in the electronic structure, and for thin films or multilayer systems where interference effects of the visible light between various interfaces have to be taken into account.

In this paper, we demonstrate the feasibility of applying SXMCD to the investigation of the magnetic properties of ultrathin films by measuring the temperature dependent SXMCD of Ni films of various thicknesses grown on Cu(100), yielding layer thickness dependent Curie temperatures.

2. Experimental

The experiments were performed at the AT&T Bell Laboratories Dragon high resolution soft-X-ray beamline located at the National Synchrotron Light Source, Brookhaven National Laboratories [1]. The beamline optical arrangement in the circular polarization mode has been described previously [2]. In the MCD experiments presented here, the monochromator resolution was kept at 0.5 eV in the 840–890 eV photon energy range and the degree of circular polarization was calculated to be $\approx$ 90%. The X-ray absorption spectra from Ni on Cu(100) films were recorded in the partial electron yield mode where electrons with kinetic energies higher than 20 eV are counted using a channeltron. The light was at grazing incidence along the (010) surface plane direction and the illuminated area of the sample was 9 × 2 mm^2. A remnant magnetic field was established in the Ni film by using an electronmagnet pulsed parallel or anti-parallel to the same (010) surface direction. For the temperature dependent MCD experiment, the absorption intensity was measured only at the L_3 white line and at 5 eV below. This allows about 20 cycles of parallel and anti-parallel magnetizations in 5 minutes to achieve a $\approx$ 0.3% sensitivity in the MCD to absorption ratio. This sensitivity is mainly limited by the instability of the electron orbit of the synchrotron radiation ring.

3. Results and discussion

Thin films of Ni were evaporated onto a clean and well-ordered Cu(100) surface which was kept at room temperature. During evaporation a beam of 2 keV electrons was directed onto the sample, and the electron beam induced crystal current (EBIC) was monitored [6]. Changes in the current can be related to abrupt changes in the Ni/Cu Auger intensity, which in turn can be associated with the completion of a monolayer [7,8]; the thickness of the Ni films are determined by counting the number of oscillations, in a way similar to the reflection high energy electron diffraction (RHEED) method used in metal molecular beam epitaxy (MBE) [9]. Fig. 1 shows such oscillations and suggests good epitaxy growth for up to 6 layers [8,10]. For the 6 layer sample, the Cu 2p X-ray photoemission spectroscopy (XPS) signal is an order of magnitude smaller than that of the Ni 2p as expected. No changes in the XPS spectra were found when annealing the sample up to 500 K, suggesting the absence of island formation or alloying. We note that no EBIC oscillations were observed when the Cu substrate was kept at 400 K during evaporation, and that the corresponding Cu 2p XPS intensity
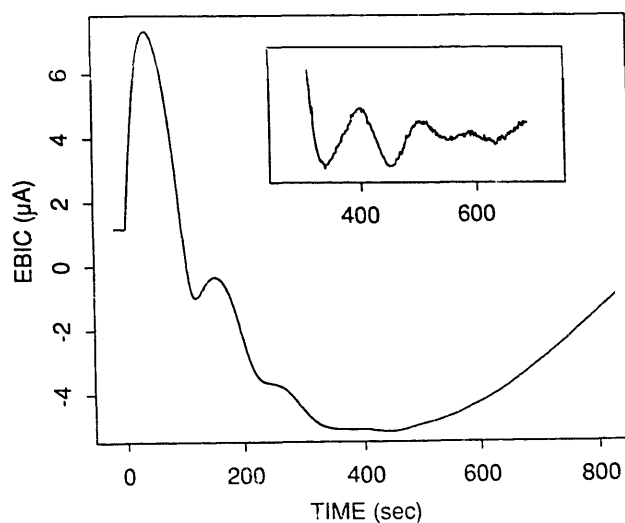
Fig. 1. Electron beam induced crystal current (EBIC) as a function of Ni evaporation time. The Cu(100) substrate was kept at room temperature. The inset shows a 20 times enlargement of the spectrum with a smooth background subtracted.

was comparable to the Ni 2p, strongly indicating the formation of Ni islands. Evaporation at 77 K substrate temperature gave only 2 oscillations but showed that same XPS as for the room temperature evaporation, indicating that flat epitaxial films cannot be grown beyond 2 layers due to insufficient Ni surface mobility, resulting in statistically rough surfaces. The base pressure of the vacuum chamber was in the low 10^{-10} Torr. The carbon and oxygen contamination measured by XPS 4 hours after the evaporation is found to be less than 0.1 monolayer.

Fig. 2 shows the X-ray absorption spectrum (XAS) and MCD spectrum of a bulk like thick Ni film. A recent relativistic Slater–Koster tight-binding analysis [11] has obtained a good fit for most of the spectral features in both the XAS and MCD spectra, and yielded values for the Ni 3d spin–orbit interaction, exchange splitting and local magnetic moment. It is interesting to note that while peaks A, A' only appear in the XAS spectrum and can be reproduced within the tight-binding analysis, peaks B, B' are imperceptible in the XAS spectrum, but manifest as distinct shoulders in the MCD spectrum. The latter peaks can not be explained within the tight-binding analysis, indicating the need to include many body effects [12].

The MCD signal for various Ni film thicknesses as a function of temperature a. shown in fig. 3. The data were taken while the sample was warming up. Note the striking dependence of the Curie temperature on film thickness. The solid curves in fig. 3 are fits using the assumption that the magnetization is proportional to $(T_c - T)^\beta$, with T_c being the Curie temperature. Limited by our current MCD detection sensitivity ($\approx 0.3\%$), it is very difficult to measure the small MCD signal near the Curie temperature with high accuracy, and therefore, a reliable β parameter can not be obtained at this time. The Dragon beamline is about to be upgraded to a double-headed version to enhance the current MCD detection sensitivity by two orders of magnitude [13]. Fig. 4 shows the thickness dependence of the Curie temperature, together with the results of Ni films on Cu(111) from ref. [5]. It can be observed that the Curie temperature for a given thickness is
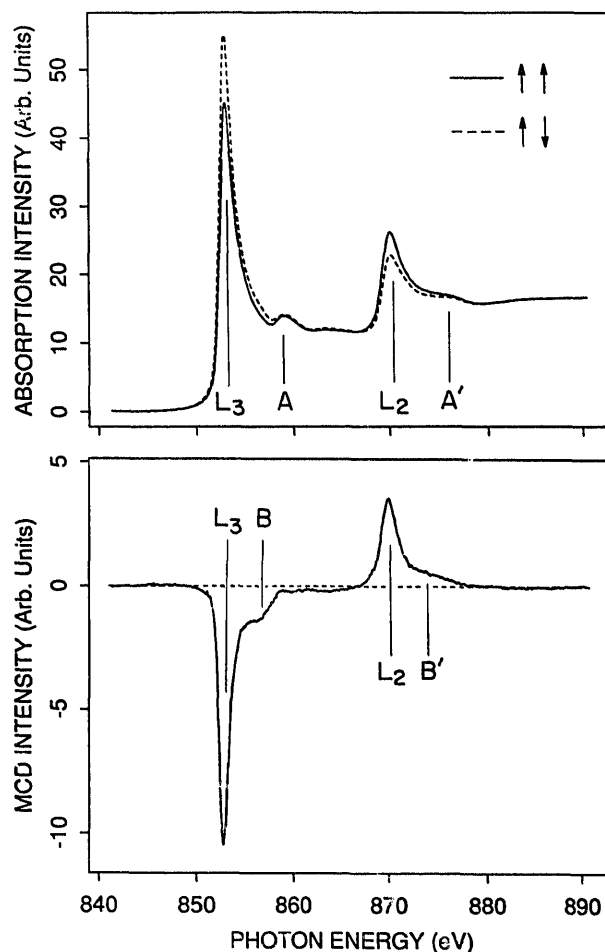

Fig. 2. $L_{2,3}$ X-ray absorption spectra (XAS) and the magnetic circular dichroism (MCD) of a thick Ni film taken with the spin directions of the photons and that of the majority 3d electrons parallel and anti-parallel to each other. Peaks observed at 853 and 871 eV are the L_3 and the L_2 white lines. The sum of the average L_3 XAS peak height is set at 100. The MCD spectrum shown in the power panel is obtained from the difference of the two XAS spectra shown in the upper panel. Fine features labeled as A, A' and B, B' are discussed in the text.

substantially lower for Ni on Cu(100) than on Cu(111). Moreover, extrapolating down to lower coverages, two monolayers of Ni on Cu(100) would have a Curie temperature of about 50 K and one monolayer of Ni might be non-magnetic, unlike the Ni on Cu(111) case. This would then contradict the results from theoretical calculations [14,15] and suggestions from experiments using electron capture spectroscopy [16] and spin-polarized angle-resolved photoemission [17]. Ear-
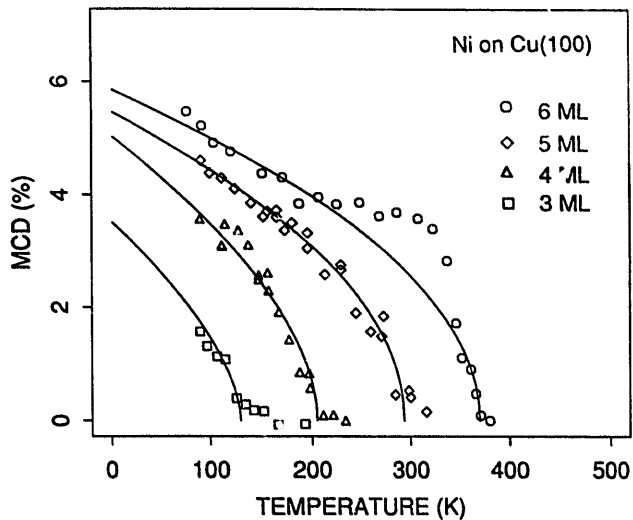
Fig. 3. Magnetic circular dichroism (MCD) signal at the L_3 white line from Ni films of various thickness (in units of monolayers) on Cu(100) as a function of temperature. Solid curves are power law fits as described in the text.

lier spin-polarized photoemission [18], anomalous Hall effect [19] and superconducting proximity effect [20] measurements show some possible magnetization effect for one monolayer Ni on Cu. Since these works did not specify the epitaxy or the Cu substrate orientation, no conclusion could be drawn for Ni on Cu(100). Results from magnetization measurements of Ni layers sandwiched between Cu(100) layers show a clear magnetiza-
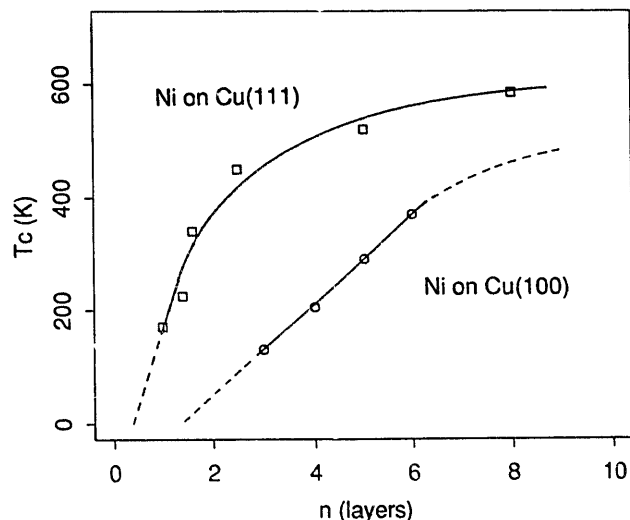
tion for two layers but are rather ambiguous for one monolayer [21]. We note deviations of the MCD temperature dependence from the fitted power law behavior at around 330 K in the 6 Ni layer case. It is not clear at this time whether or not this effect is related to possible Ni surface reconstructions.

To conclude, we have measured the magnetic properties of ultrathin Ni films on a Cu(100) substrate using soft-X-ray magnetic circular dichroism spectroscopy, thereby demonstrating the feasibility of using this technique on a broad range of magnetic surfaces and ultrathin films, particularly, on multicomponent systems. Our data seem to suggest that a monolayer of Ni on Cu(100) is non-magnetic.

Acknowledgements

References

[1] C.T. Chen, Nucl. Instr. and Meth. A 256 (1987) 595;
C.T. Chen and F. Sette, Rev. Sci. Instr. 60 (1989) 1616.
[2] C.T. Chen, F. Sette, Y. Ma and S. Modesti, Phys. Rev. B 42 (1990) 7262;
F. Sette, C.T. Chen, Y. Ma, S. Modesti and N.V. Smith, in: X-ray Absorption Fine Structure, ed. S.S. Hasnain (Ellis Horwood Pub. Co., New York, 1991).
[3] G. Schütz, W. Wagner, W. Wilhelm, P. Kienle, R. Zeller, R. Frahm and G. Materlik, Phys. Rev. Lett. 58 (1987) 737;
G. Schütz, R. Wienke, W. Wilhelm, W. Wagner, P. Kienle, R. Zeller and F. Frahm, Z. Phys. 75 (1989) 495;
G. Schütz, in: X-ray Absorption Fine Structure, ed. S.S. Hasnain (Ellis Horwood Pub. Co., New York, 1991).
[4] P.A. Snyder and E.M. Rowe, Nucl. Instr. and Meth. 172 (1980) 345;
J.C. Sutherland, P.C. Keck, K.P. Griffin and P.T. Takacs, ibid 195 (1982) 375;
U. Heinzmann, Phys. Scr. 17 (1987) 77;

Fig. 4. Thickness dependence of the Curie temperature T_c of Ni on Cu(100) (this work) and that of Ni on Cu(111) (ref. [5]).

J. Garbe and J. Kirschner, Phys. Rev. B 39 (1989) 9859;
T.C. Van Cott et al., J. Phys. Chem. 93 (1989) 2999.

[5] C.A. Ballentine, R.L. Fink, J. Araya-Pochet and J.L. Erskine, Appl. Phys. A 49 (1989) 459; Phys. Rev. B 41 (1990) 2631.

[6] L.H. Tjeng, R.W. Bartstra and G.A. Sawatzky, Surf. Sci. 211 & 212 (1989) 187.

[7] C. Argile and G.E. Rhead, J. Phys. C 30 (1982) L193;
M.-G. Bartthes-Labrousse and G.E. Rhead, Surf. Sci. 116 (1982) 217;
G.E. Rhead, Contemp. Phys. 24 (1983) 535;
D. Chadwick, M.A. Karolewski and K. Senkiw, Surf. Sci. 175 (1986) L801.

[8] Y.U. Idzerda and G.A. Prinz, (in preparation).

[9] S.T. Purcell, B. Heinrich and A.S. Arrott, Phys. Rev. B 35 (1987) 6458.

[10] M.A. Abu-Joudeh, B.M. Davies and P.A. Montano, Surf. Sci. 171 (1986) 331.

[11] C.T. Chen, N.V. Smith and F. Sette, Phys. Rev. B 43 (1991) 6785.

[12] T. Jo and G.A. Sawatzky, Phys. Rev. B 43 (1991) 8771.

[13] C.T. Chen, F. Sette and N.V. Smith, Appl. Opt. 29 (1990) 4535.

[14] C.S. Wang, A.J. Freeman and H. Krakauer, Phys. Rev. B 24 (1981) 1126.

[15] J. Tersoff and L.M. Falikov, Phys. Rev. B 26 (1982) 6186.

[16] C. Rau and S. Eichner, J. Magn. Magn. Mater. 35 (1983) 25.

[17] M.A. Thompson and J.L. Erskine, Phys. Rev. B 31 (1985) 6832.

[18] D.T. Pierce and H.C. Siegmann, Phys. Rev. B 9 (1974) 4035.

[19] G. Bergmann, J. Magn. Magn. Mater. 35 (1983) 68.

[20] J.S. Moodera and R. Mesevey, Solid State Commun. 59 (1986) 295.

[21] L.R. Sill, M.B. Brodsky, S. Bowen and H.C. Hamaker, J. Appl. Phys. 57 (1985) 3663.