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of bulk as well as thin-layered ferromagnets.

The magnetization of a very thin magnetic layer (down to an atomic plane) can be measured, provided it is embedded in a nonmagnetic matrix.

1. INTRODUCTION

Scattering of x-rays at grazing incidence is providing exciting new information on the composition and density fluctuations at the surface. The measurement of the intensity reflected for different angles of incidence θ_i gives the depth profile of the scattering density. [1] The diffuse scattering around the specular beam determines the critical fluctuations at surface phase transitions, as in smectic/nematic liquid crystals.[2] The measurement of the surface Bragg reflections in conditions of grazing incidence provides the structure at the surface and its relation to that of the bulk.[3] For all these problems, the great advantage of x-rays in comparison with more surface-sensitive radiations is the capability of interpreting accurately and unambiguously the intensities measured. The sensitivity of x-rays to surface phenomena is enhanced in the grazing incidence geometry. Even so, the scattered intensities are weak, because the number of the

scattering centers in the surface layer is small: hence powerful sources are needed.

The information that neutrons can provide is in principle parallel to that obtained by x-rays. As for conventional diffraction, neutrons are to be preferred to x-rays when dealing with elements with a favorable scattering amplitude (as hydrogen) [4], and whenever magnetic phenomena are involved [5]. The present paper deals primarily with the magnetic aspects of the scattering of neutrons at grazing incidence, and in particular, concentrates on the most simple study that can be made in such a geometry--the study of the intensity of the reflected beam.

2. NEUTRON REFLECTIVITY

A detailed description of the reflectivity of slow neutrons from surfaces has already been presented in the literature [5-7], and only a brief account will be given here. The neutron interaction with a material can be described in terms of a refractive index n , which may change as a function of the distance from the surface z .

If the material is magnetized parallel to the surface, neutrons polarized parallel (+) or antiparallel (-) to the direction of the applied magnetic field H have refractive indices:

$$n^{\pm}(z) = 1 - \frac{\lambda^2}{2\pi} \left[\frac{b}{v} \pm c [B(z) - H] \right] \quad (1)$$

where λ is the neutron wavelength, b the average nuclear scattering amplitude in the atomic volume v , c is a constant, and $B(z)$ is the magnetic induction in the material. The refractive index for neutrons, for wavelengths of the order of a few Angstroms, is only slightly different from unity, since for all materials $|b/v| < 10^{-5} \text{ \AA}^{-2}$. However, the constant $c = 2\pi\mu_N m/h^2 = 2.3 \times 10^{-10} \text{ \AA}^{-2} \text{ Oe}^{-1}$: even modest magnetic fields are sufficient to yield a sizeable spin-dependence of $n-1$. The perpendicular component of the neutron momentum in vacuum, $k_0 = 2\pi \frac{\sin \theta_1}{\lambda}$ is modified by the refractive index in the material to:

$$n^{\pm}(z) \cdot k^{\pm}(z) = \sqrt{k_0^2 - 4\pi \left(\frac{b}{v} \mp c [B(z) - H] \right)} \quad (2)$$

The reflectivity for a given material is solely a function of k_0 ; $R^{\pm}(k_0)$ are optical (and nonlinear) transforms of $B(z)$. Practically speaking, $R^{\pm}(k_0)$ are scanned by keeping the angle of incidence θ_1 with the surface fixed and varying the neutron wavelength up to the limit for which $k^{\pm}(z) = 0$ for all z 's: at which point the reflectivity is total.

Except in very particular cases, the reflectivity cannot be given in simple analytical form. However, its dependence from the magnetic induction can be given in an approximate expression for $R \ll 1$, $cB \ll b/v$;

$$R^{\pm} \sim R_n \left[1 \pm c_1 \int_0^{\infty} \frac{dB(z)}{dz} \exp(2 k_n z) dz \right] \quad (3)$$

where R_n , and k_n are respectively the reflectivity and the perpendicular component of the neutron momentum in the material for $B=0$. Within the given approximations $\Delta R = R^+ - R^-$ is a Fourier transform of the derivative of the magnetic profile. In practice, however, the full reflectivity curves are numerically calculated starting from model magnetization profiles.

3. THE INSTRUMENT

The layout of the prototype, set up at the Intense Pulsed Neutron Source at Argonne, is presented in Fig. 1. Neutrons emitted from a uranium target struck by protons are slowed down by a grooved moderator of solid methane. The slowed down neutrons have a burst width of 180 μ sec; the repetition rate of the source is 30 pulses/sec. The hot tail of the spectrum is eliminated by passing the beam through a cold

(78K) beryllium filter, which effectively eliminates the neutrons of wavelengths less than 3.8 Å.

The polarization efficiency of the Fe-Ag supermirror [8] is 96% for the neutron wavelength range of 4-8 Å. The efficiency of the non-adiabatic neutron flipper [9] is close to unity. The neutrons are collected by a small (2.5 cm diameter) area detector with a resolution element of approximately 1 x 1 mm, set at a distance of 60 cm from the sample. The detector consists of a 0.5 mm thick Li^6 glass scintillator in front of the photocathode of a multichannel plate, followed by an x-y resistive readout [10]. Each pixel, of 0.4x20 mm area, is separately analyzed by time-of-flight, with a channel width of 50 μs .

The sample surfaces have typically an area of 15x50 mm. The surfaces have good reflecting properties if they have a mirror finish, and if they are flat within one optical fringe. Rather than polishing ingots, in most instances it was found easier to obtain good quality samples by sputtering or vapor depositing the material (for a thickness of a few microns) onto a polished substrate made of a weakly reflecting solid, such as silicon.

A measured reflectivity profile is presented in Fig. 2, and compared with the calculated reflectivity. In the calculation, the reflectivity is averaged over the angular spread of the neutron beam, which is assumed to have Gaussian shape, and it is further modified to take account of the microroughness of the sample surface. This is assumed [11] to modify the reflectivity as a Debye-Waller factor

$$I(k_o) = I_o(k_o) \exp - [4 k_o k_n \langle z_D^2 \rangle] \quad (4)$$

The parameter $\langle z_D^2 \rangle^{1/2}$ which represents the surface roughness, is obtained by fitting the calculated with the experimental reflectivity. We will now discuss some applications.

4. SUPERCONDUCTORS AND FERROMAGNETS

An external magnetic fields H deeply perturb the Meissner state of superconductor close to the surface, for a thickness of several hundred Angstroms. If H is lower than a critical value H_{c1} , the bulk of the material is still superconducting ($B=0$) but the magnetic field penetrates the surface with an exponential decay defined by a penetration depth Λ . This was measured [6] in niobium (a type II

superconductor) at different temperatures and in different magnetic fields, an extrapolated value at $T=0$ of $\lambda = 410 \pm 40 \text{ \AA}$ was obtained. The sharp crossover from the superconducting to the normal state above H_c has been observed [7] in pure lead, a type I superconductor. Alloying minute amounts (0.8%) of bismuth with lead, a material has been obtained with an incipient type II behavior. For this alloy the neutron results indicate [7] a much smoother transition from the superconducting to the normal state, in agreement with the expected presence of an intermediate magnetic phase.

A surface sheath of superconductivity has been predicted [12] to occur between H_{c2} (the critical field, at which the bulk becomes normal) and $H_{c3} \approx 1.7H_{c2}$. The diamagnetism of this surface sheath is expected [13] to be strongest for an incipient type II superconductor (like the Pb-08% Bi alloy, for which $H_{c1} \sim H_{c2}$). The ratio of the spin-dependent reflectivities in the two regimes is presented in fig. 3. The magnetic signals for $H < H_{c1}$ and for $H_{c2} < H < H_{c3}$ have different shapes. A preliminary analysis of the data has provided the tentative magnetic profiles presented in fig. 4.

For simple metallic ferromagnets, the bulk

magnetization is perturbed in proximity of the surface boundary. In the ground state the perturbation is expected to be limited to a few outer atomic planes. However, at finite temperatures, the surface magnetization is perturbed over a correlation depth which becomes infinite at the Curie point. Close to T_c , both the surface and the bulk magnetization vary as $M \propto (1 - T/T_c)^\beta$, but the surface critical exponent β_s is different from the β of the bulk. The two solutions join at a correlation depth [14] which is also a function of β_s ; hence the latter quantity can be determined by magnetization profiles. An experiment is at present underway in order to determine the surface critical properties of metallic nickel.

The magnetization close to the surface may also be changed as a result of a mechanical or a thermal treatment. For instance, an experiment [15] was done on a film - 2600 Å thick - of ferromagnetic iron oxide (nominally Fe_3O_4). The magnetization was found uniform across the film; however, after this was roasted in oxygen in order to improve its magnetic remanence, a magnetic dead layer appeared at the surface, for a thickness of ~150 Å.

5. LIMITS OF RESOLUTION

The general purpose of the technique is to measure a perturbation of the refractive index Δn which occurs over a depth Δz . Let us see what should be the range of the measured reflectivity in order to determine Δz . If the perpendicular component of the neutron momentum in the medium is k_n , the perturbation Δn of depth Δz gives rise to an interference maximum at $k_n = \frac{\pi}{\Delta z}$. The (unperturbed) reflectivity for this value of k_n is $R = \left| \frac{2}{\pi} \frac{b}{v} (\Delta z)^2 \right|^2$, where b represents the average scattering amplitude of the medium. Numerically $R < 10^{-10} (\Delta z)^4$ (Δz expressed in Angstroms): a Δz of 100 Å is easily determined while a Δz of 10 Å requires measurements down to a region in which the unperturbed reflectivity is 10^{-6} of the incident beam. This feat is quite difficult to achieve with present-day neutron sources.

If the depth Δz of a perturbation Δn of the refractive index already is known by other means, neutron measurements over a limited range of k_n can still determine Δn , particularly if this is spin-dependent. In fact, the total magnetization of an extremely thin magnetic layer (down to one atomic plane) can be measured, especially if the

layer is not at the surface, but embedded in a nonmagnetic matrix.

Suppose that we have a magnetic layer of thickness d_2 , and with refractive index characterized by B , b_2/v_2 ; the layer is deposited on a nonmagnetic support of unitary scattering amplitude b_1/v_1 , and covered by thickness d_1 of the same material. If d_2 is small, the expression of the reflectivity can be expanded in powers of d_2 . The ratio of reflectivities for the two neutron spin states, containing only terms linear in d_2 , is:

$$\frac{R^+}{R^-} = 1 + \frac{8 d_2 B k_0}{b_1/v_1} \sin [2 d_1 k_1] \quad (5)$$

Figure 5 shows the spin dependent reflectivities for a layer of Fe ($2.2 \mu_B/\text{atom}$), $15 \text{ \AA}$ thick, deposited on gold and covered with an overlayer of gold $200 \text{ \AA}$ thick. The sine term appearing in eq. (5) is an enhancing factor: if no overlayer were present, the spin dependence of the cross section would appear only in higher powers of d_2 , which have been neglected in Eq. (5).

Preliminary experiments have basically confirmed the validity of the above treatment, although the sample surfaces used up to the present have not had a reflectivity of quality comparable to that

presented in fig. 2. As observed in Eq. (5) the magnetic quantity that is determined in the experiment is $B \cdot d$, or the total magnetization of the sample. However, an inherent assumption in formulating to Eq. (5) is the planar nature of all layers - magnetic and nonmagnetic alike. Only in this geometry does the incident neutron plane wave split in two equally plane refracted and reflected waves. If the magnetic layer is laterally modulated (for example, if it is not a plane at all but an assembly of droplets, or if magnetic fluctuations are present) scattering of the neutrons would occur outside of the reflected beam. Thus a reflectivity profile is capable of determining if the magnetic layer is indeed flat. On the other hand, even if this layer is ferromagnetic [16] around a characteristic temperature critical fluctuations would give rise to a diffuse scattering around the reflected beam. While the current scattering theories [17-19] do not deal in detail with the case of submerged layers, it seems reasonable that even the diffuse magnetic signal is enhanced as the reflected intensity; this would make feasible the study of the critical fluctuations in a truly two-dimensional magnetic system.

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FIGURE CAPTIONS

Fig. 1 Polarized neutron reflectometer. A pulsed beam of cold neutron ($\lambda > 3,8\text{\AA}$) ions polarized by reflection of a magnetized mirror. The polarized beam is partially reflected by the magnetized sample surface (magnetic fields normally plane of the page). Reflectivity measurements for the two states of the neutron spins are taken in rapid alternation, by switching the spin flipper between neutron pulses.

Fig. 2 Reflectivity of a Pb film, one micron thick, vapor deposited on a silicon single crystal. The measurements were taken at room temperature. The calculation reflectivity is for a glancing angle $\theta_i = 0.358 \pm 0.013^\circ$, and for a surface roughness $\langle z^2 \rangle^{1/2} = 58\text{\AA}$.

Fig. 3 Ratio of the R^+/R^- reflectivities for the Pb-08% Bi at 5.7K in the bulk superconducting state (300 Oe) and in the state of surface superconductivity (540 Oe). The curves represent the preliminary fitting with the data. Angle of incidence $\theta_i = 0.358 \pm 0.015^\circ$.

Fig. 4 Tentative magnetic depth profiles for the Pb-08% Bi film. These profiles correspond to the curves drawn in Fig. 3.

Fig. 5 Calculated reflectivities for neutrons polarized parallel (+) and antiparallel (-) to the ferromagnetic moments ($2.2 \mu_B/\text{atom}$) of a film of iron, 15 Å thick. The iron is deposited on gold, and covered with an overlayer of gold 200 Å thick. Angle of incidence $\theta_i = 0.480 \pm 0.015^\circ$.