Density Measurements: Rutherford Backscattering Spectroscopy

1 Introduction

As part of the AXAF Reflectivity Study, it is of interest to perform density measurements on the films used to coat the mirrors being investigated. The interest in density, ρ, derives from the fact that the critical angle for reflectivity, θ_c, scales as $\rho^{1/2}$. The critical angle is inversely related to energy (through the optical constants) so that, for a chosen angle of operation (*i.e.* for a fixed grazing angle associated with a particular mirror design) the critical angle determines the high energy cutoff of operation. To maximize the upper energy range, then, it is important to achieve coating densities that are as high as possible.

Experimental measurements of reflectivity generally yield results which are lower than those predicted by theoretical calculations in which densities equal to the bulk density of the coating material are assumed. Thus, it is of interest to measure densities for coatings in an effort to correlate density with x-ray reflectivity.

2 Theoretical Aspects

The technique for determining coating density using Rutherford Backscattering Spectroscopy (RBS) involves bombardment of the coated sample with α-particles. The fraction of incident particles backscattered into a given annular region is directly proportional to the "areal density" η defined by

$$\eta = \int_0^t \rho(x)\ dx = \bar{\rho} t \tag{1}$$

where $\bar{\rho}$ is the mean density and t is the coating thickness (see below for a more thorough discussion of this relationship). Thus, determination of $\bar{\rho}$ requires an independent measurement of the coating thickness. For purposes of the AXAF Reflectivity Study, where possible, thickness measurements will be made with a Scanning Transmission Electron Microscope (STEM) by viewing cross-sectional samples of the coating prepared from silicon wafer chips coated with the mirrors. Where STEM measurements can not be performed, thickness measurements will be performed by surface profilometry. The thickness measurements are accurate to $\sim$20 Å with the STEM and to $\sim$50-75 Å by profilometry. The uncertainty in the areal density measurement by RBS is about 1%; hence, for thin coatings, the accuracy to which the thickness can be measured determines the uncertainty in the final density determination. It should be noted that this technique yields an average density for a given coating. X-ray reflectivity measurements are sensitive to densities averaged over the associated skin depth δ which is a function of the x-ray energy and grazing angle (see Figure 1):

$$\delta = \frac{\hbar c}{\gamma} \frac{\xi}{E} \tag{2}$$

Figure 1: X-ray skin depth as a function of energy for gold coatings at a grazing angle of 30 arcmin.

where

$$\xi^2 = \frac{1}{2}\left\{\sin^2\theta - \alpha + \left[\left(\sin^2\theta - \alpha\right)^2 + \gamma^2\right]^{1/2}\right\}. \tag{3}$$

Here α and γ are derived from the complex dielectric constant $\kappa = 1 - \alpha - i\gamma$. Thus, the densities inferred from the RBS measurements (in conjunction with thickness measurements, which also represent an averaging over a somewhat "bumpy" surface) may not provide the full information required to predict x-ray reflectivity behavior. Still, the mean density is clearly a quantity that should be maximized for good overall reflectivity, and thus the measurements have merit. In addition, the measurements provide an indication of how successfully one can obtain bulk material densities by thin film techniques.

A typical set-up for RBS measurements is shown in Figure 2. A beam of α-particles is accelerated to several MeV, collimated, and directed toward the sample film. A solid state detector is used to detect the backscattered beam at some angle θ. In practice, a sample holder containing up to 20 samples is placed at the target position; individual samples are chosen for investigation by properly adjusting the position of the sample holder. Typical beam sizes, after collimation, are ~ 1 mm.

The number of particles scattered into a cone bounded by θ and $\theta + d\theta$ is given by

$$\frac{dN(\theta)}{I} = \left(\frac{\pi D^2 N_0}{8A}\right)\left[\frac{\sin\theta\ d\theta}{\sin^4\left(\frac{\theta}{2}\right)}\right]\eta \tag{4}$$

Figure 2: Schematic diagram of set-up for Rutherford Backscattering Spectroscopy measurements.

where

$$D = \frac{zZe^2}{4\pi\epsilon_0 E}. \tag{5}$$

Here I is the incident intensity of α-particles (of charge z and energy E), Z and A are the atomic number and atomic mass of the scatterer, and N_0 is Avogadro's number. Thus, as noted above, for a fixed target material the fraction of particles scattered into a particular annulus is proportional to the areal density η.

Given that, in the bombardment of a thin film coating with energetic α-particles, considerable penetration into deeper layers (such as the undercoating or substrate material) may occur, it is necessary to identify which scattered particles are associated with the material under investigation. This may be accomplished by considering the kinematics of the elastic collisions. The ratio of scattered energy to incident energy for the α- particles is dependent upon the scattering angle θ and the mass m_t of the target scatterer:

$$\frac{E_{sc}}{E_0} = \left[\frac{\left(m_t^2 - m_\alpha^2 \sin^2\theta\right)^{1/2} + m_\alpha \cos\theta}{m_t + m_\alpha}\right]^2. \tag{6}$$

Thus, for a monoenergetic input beam, the energy of the particles scattered from a given target material is well defined; energy discrimination can be used to identify and count only the scattered particles of interest. Of course, both the finite angular size and the finite energy resolution of the detector will result in a distribution of energies associated with a particular scattering material. Furthermore, the finite thickness of the coating layer will broaden the energy distribution due to $\frac{dE}{dx}$ losses associated with inelastic collisions between the α-particles and electrons; the thicker the coating, the larger the broadening of the energy spectrum. Separation of adjacent energy peaks in the spectrum may become difficult for certain combinations of scattering materials. In general, such resolution difficulties can be circumvented by increasing the energy of the α-particles.

3 Experimental Technique

The procedure for determining areal density by RBS involves a comparison of the coating scattering results with those from a sample with a known areal density. Such samples are produced by ion implant whereby the number of ions deposited on a sample are determined by a charge sensitive monitor; the implant is confined to a small area and is uniform over this area so that the areal density may be calculated. The standard sample used for comparisons in this study consists of an implant of gold into silicon with an areal density of 2×10^{15} cm^{-2}. A scattering run gated for a fixed number of incident α-particles (actually, a fixed integrated charge - typically, 50 μC) yields the calibration number of counts in a fixed detector subtending some known fraction of an annular region. Energy spectroscopy of the α-particles scattered into this annulus allows discrimination of particles scattered from different elements (*e.g.* scattered from the silicon substrate rather than the gold). For each sample, then, a comparison run is made, gated for the same number of input ions. The ratio of counts in the gold peak from the sample to that from the calibration source is equal to the ratio of areal densities:

$$\frac{N}{N_{cal}} = \frac{\eta}{\eta_{cal}}. \tag{7}$$

For typical measurements (*e.g.* gold/chromium on silicon), 2 MeV α-particles are used for the scattering. This allows clear separation of peaks from the silicon substrate, the chromium undercoat, and the gold (or iridium) coating (see Figure 3). For some samples, this peak separation is not possible at 2 MeV (*e.g.* AuPd) and operation at 3 MeV is necessary.

As mentioned above, coating thickness plays a role in the energy distribution of the scattered α-particles. Thus, while the high energy side of the distribution is well-defined, the low energy side is determined by the $\frac{dE}{dx}$ losses associated with penetration into the coating. This is illustrated in Figure 4 which shows comparison profiles for two coatings of different thicknesses. In Figure 4(a), results from a sample coated with $\sim$ 300 Å of gold over a thin layer of chromium are plotted. The broad region associated with the substrate is more complicated than that shown in Figure 3 due to the presence of a silicon-oxide layer on the substrate. In Figure 4(b), results from a $\sim$ 700 Å gold coating are shown. Note that the upper-edge of the gold peak is coincident for the two plots, as expected, but that the lower-edge is pushed to lower energies for the thicker coating due to penetration losses. Similarly, the entire chromium peak is shifted toward lower energies for the thicker coating because

Figure 3: RBS output showing peaks associated with scattering from gold coating, chromium undercoating, and silicon substrate. Events in the gold peak are used for comparison with a calibrated sample in order to determine the area density.

complete penetration of the gold layer is required for any scattering off of the chromium layer. Of course, this effect can be seen in the profile associated with the substrate as well. Such a thickness effect can, of course, be used to estimate the actual coating thickness; such estimates require the coating density as input, however, and are thus not appropriate for cases in which the density is not known. Note that the low energy end of the substrate profile is coincident for the two samples; this hard cut-off represents an energy threshold for the detector.

4 Preliminary Results and Conclusions

The scattering results for a number of samples measured in a test run are listed in Table I. The first column lists the sample mirror associated with the measurement (recall that it is not the mirror itself measured, but rather a witness sample coated with the mirror). The second column indicates the coating type (gold, nickel, etc.) while the third column indicates the nominal thickness of the coating. More accurate results for the thickness will become available upon completion of the STEM investigation of the samples.

Figure 4: Scattering profiles for a)$\sim$ 300 Å and b)$\sim$ 700 Å of gold over a thin chromium layer, atop a silicon substrate (with a thin silicon-oxide layer). Effects of energy loss due to finite coating thickness can be seen, as explained in text.

Table I. RBS Results for Deposition Effects Samples

Sample	Integration Region	Net Counts	ρt (atoms cm^{-2})	ρt (gm cm^{-2})	t (Å) (α- step)	ρ (g cm^{-3})
CT#1	770-811	8624	2×10^{15}	6.54×10^{-7}		
DE1	754-820	818334	1.90×10^{17}	6.22×10^{-5}	525	11.85
DE2	750-822	878102	2.04×10^{17}	6.68×10^{-5}	490	13.63
DE3	746-822	782028	1.81×10^{17}	5.92×10^{-5}	547	10.82
DE4	759-824	559315	1.30×10^{17}	4.25×10^{-5}	590	7.20
DE5	759-822	627542	1.45×10^{17}	4.74×10^{-5}	517	9.17
DE6	759-822	609584	1.41×10^{17}	4.61×10^{-5}	370	12.46

There are several factors associated with the measurement process which may cause systematic errors in the density determinations outlined above. Thus, the numbers presented should be considered very preliminary; further analysis, and quite possibly remeasurement, will be required to determine realistic values.

- Improper beam alignment can cause only partial illumination of the sample. In this case, a lower density would be inferred in that the incoming flux would still be counted, but some fraction would miss the sample thus yielding a smaller number of scattered particles than expected. Such an effect can generally be identified in that the particle beam leaves a slight mark on the sample. Thus, it can be determined whether or not the beam was striking the sample close to an edge.

- Normalization difficulties occur if the beam monitor is not used. This did occur for some of the acquired data because the monitor was being repaired. Typically, one merely times successive runs and normalizes the data based upon a comparison of the counts in the substrate region of the profile. However, since different coating thicknesses can push this distribution toward lower energies, and since the lower end cut-off is instrument related (*i.e.* it is the same regardless of the coating thickness) the actual number of counts in the two regions should *not* be identical for identical input beam fluxes. Samples obtained while operating in this mode will be remeasured.

- An overall uncertainty exists which is associated with the uncertainty in the areal density of the calibration sample. This uncertainty is not known for the current sample; procurement of a calibration sample for the AXAF reflectivity is desirable so that comparisons can be made with a sample for which documentation can be obtained.

- Measurements made on coatings other than gold are difficult to assess. Since only a gold calibration sample exists, the areal density associated with samples other than gold must be inferred based upon the ratio of densities of the various materials to that of gold. However, since it is the densities that are being investigated, this process contains an ambiguity. The disirable mode of operation would be to obtain calibration standards for each of the coating materials under investigation.