WAVELET INVERSE NEUTRON SCATTERING STUDY OF LAYERED METALLIC NiC-Ti COMPOSITES

G. Saab[1], B. DeFacio[2], H. Kaiser[3], and D.L. Worcester[4]
[1]Dept. of Physics and Astronomy and Medical School
[2]Dept. of Physics and Astronomy
[3]Missouri University Research Reactor
[4]Missouri University Research Reactor, Division of Biological Sciences
Missouri University
Columbia, MO 65211

INTRODUCTION

Composites are prevalent in high technology devices such as aircraft, computers, automobiles and communications systems. They improve brittleness and provide a lower density which enhances mechanical strength. Electron and light manipulating composites will be used more and more in the future. It is necessary to have a capability of inspecting composites, both to assure production quality and as a baseline for later NDE. In this paper, we present a study using wavelet, inverse neutron optics and the grazing angle neutron spectrometer, GANS, at the Missouri University Research Reactor, MURR.

Neutrons are electrically neutral matter-waves with mass just greater than the proton. They have good penetration depths since they scatter almost entirely from the nuclei in a solid. This property gives thermal neutrons the capability of measuring the amount of residual stress in many solids [1,2] by diffracting from the atomic planes. However, since the 10 MWatt reactor provides the 10^{14} neutrons/cm^2 sec incident onto the collimeter it is not a technique for all applications in NDE. But it reaches deeply into the solid and relays detailed information on an atomic length scale [3] so it may be worthwhile to learn what can (and what cannot) be determined from this modality.

ANALYSES

The theoretical description used has deep roots in physics going back 50 years to Fermi and Zinn [4], who showed experimentally that neutrons have all of the optical properties of light: reflection, refraction, interference, diffraction, collimation, polarization and can be made quasi-monochromatic. Sears [5] and Werner and Klein [6] have written an excellent monograph and review article on neutron optics, respectively. The unpolarized neutrons satisfy the Schrödinger equation

$$\left[-\frac{\hbar^2}{2M}\Delta + V_{opt}\right]\Psi(k,\vec{r}) = E\Psi(k,\vec{r}) \tag{1}$$

where $\hbar$ is the Planck constant h divided by 2π, Δ is the Laplacian operator, V_{opt} is the effective optical potential, ψ is the quasi-monochromatic neutron probability amplitude at spatial location $\vec{r}$ and E is the energy of the neutron. The optical potential [5] is given as

$$V_{opt}(\vec{r}) = -E_n\left(n^2 - 1\right) \tag{2}$$

where the index of refraction n is related to the nuclei in the sample by

$$n = \left|1 - \frac{V_{opt}}{E_n}\right|^{\frac{1}{2}} = \left[1 - \lambda_1^2 \frac{Nb}{\pi}\right]^{\frac{1}{2}}. \tag{3}$$

In equation 3, E_n is the energy of the neutron , λ_1 is the wavelength of the neutron in the sample, N is the number density of scattering nuclei and b is the coherent scattering length of the nuclei. In a composite sample such as this one is replaced by the weighted sum of $N_i b_i$'s. It is emphasized that the optical potential describes a neutron in a many-particle system. The scattering analysis is based on Parratt [7] as extended to neutron scattering by Ankner [8], Ankner and Majkrzak [9]. An earler, complimentary analysis using this method was given in [3]. The addition of a wavelet study is presented here.

The GANS instrument at MURR was described by Kaiser et al [3], so only the parameters which are used here will be given. Several national laboratories operate similar instruments but this one is the only such spectrometer located at a U.S. University. A polarized neutron source has been added to this spectrometer since that reference appeared, but it is not used here. The sample studied has 50 layers of 5nm (50A) nominal thickness on a silicon substrate [10]. The vacuum neutron wavelength is $\lambda = 0.235$ nm with resolution $(\Delta\lambda)/\lambda \approx 1.5$ %, the transferred wave number is, $\vec{Q} = \vec{k}' - \vec{k}$, where $k' = 2\pi/\lambda'$, $k = 2\pi/\lambda$ are the final, initial wave vectors, respectively and the range of wave number is $0.02 \le Q \le 4\ \text{nm}^{-1}$. The wave number resolution range is $0.02 \le (\Delta Q)/Q \le 0.20$ and the detector is a 1/2 inch ^{3}He proportional counter.

The usual measure of an instrument such as GANS is the dynamical range of the neutron reflectance I(Q) / I(0). This instrument has a useful range of six decades. The model which describes the reflectivity measurements of specular reflection of thermal neutrons was first given for X-rays by Parratt [7] and extended to neutrons by Ankner [8,9] and others. Specular reflection is said to occur when the angle of incidence, θ , and angle of reflection, θ_R , are equal. Since $d_i >> \lambda$, a multiple scattering approach is valid but the interface profiles require more work. Let $\hat{z}$ be perpendicular to the surface of the sample and let $d_0, d_1, \ldots d_{50}$ denote the NiC / Ti interfaces. The metals diffuse into one another during the sputtering deposition but if the interface region δd_i satisfies $\delta d_i / d_i << 1$ they can be treated in two steps. Consider a collection of perfect parallel planar interfaces with incident wavenumber $\vec{k}$ and scattered wavenumber $\vec{k}'$. The wavevector transferred is $\vec{Q} = \vec{k}' - \vec{k}$. Assume that an average diffusion profile is convolved with the first profile to obtain the model profile. Let (r_i, t_i) denote the reflection and transmission coefficients at the i^{th} interface and express the reflectivity at the i^{th} and $(i-1)^{th}$ interfaces as

$$R_i = A_i^2 \left(\frac{r_i}{t_i} \right), \tag{4}$$

and

$$R_{i-1} = A_{i-1}^4 \left[\frac{R_i + F_{i-1,i}}{R_i F_{i-1,i} + 1} \right] \tag{5}$$

The phase factor, A_i, is given by

$$A_i = \exp\left(i k_j d \frac{j}{2} \right) \tag{6}$$

and the Fresnel coefficient for the (i-1, i) interface, $F_{i-1,i}$, is

$$F_{i-1,i} = \frac{k_{i-1} - k_i}{k_{i-1} + k_i} \tag{7}$$

where the refraction corrected wave vectors are

$$k_i = \sqrt{k^2 - 4\pi N_i b_i} \tag{8}$$

with k as the vacuum wave number and $N_i b_i$ are the scatterer density times the scattering lengths of the i^{th} layer. The scattering lengths, depend sensitively upon the isotope, b=b(N,Z) ie the number of neutrons N and protons Z. Thus, isotopic substitutions can solve structure problems which are otherwise ambiguous. The normal z component of the wave vector to the sample surface is

$$k = \frac{2\pi}{\lambda} \sin(\theta). \tag{9}$$

It is clear from the measured curve of reflectance vs Q that the NiC is the first layer since the data clearly shows a rounded step. Had the first layer been Ti the shape would have been given by $I(Q) / I(0) = \exp(-\alpha Q)$, with $3 / \alpha > Q > 0$. The physics behind the first peak is that of critical neutron scattering whereas the other peaks are dominated by the geometry of the 50 layers (and possibly the silicon substrate). The number of peaks and the spacings indicate that the neutrons integrate all fifty layers in direct space, z, since the uncertainty relation holds between σ_d and (Δz) as $\sigma_d (\Delta z) \geq 1$.The variances σ and (Δz) are used in the Fourier-analysis or wave-packet sense and <u>not</u> in the Heisenberg uncertainty sense. There is no sign of the Si-substrate, but the last ten layers may contain additional errors. This would presumably reduce the value of d_5 from the one given by the analysis. In Table 1 the results from a wavelet analysis using D28 wavelets is shown. The measured peaks are those shown in the figures, Q_M is the value of the wave number at the optimal location of the peak for the reconstruction to minimize the error norms from the measured peak, $\sigma_i = (\Delta Q_i)$ is the variance of the i^{th} peak and is related to the peak width in frequency domain, $d_{ij} = d_j - d_i$ is the peak separation in direct ($\vec{r}$) space and $\tilde{\sigma}_i$ is proportional to the fluctuations average width of the (ij) interface. The figures of merit are the l^2 (or energy) and l^1 (or human eye) error norms and are given by

$$l^2(\text{error}) = \left[\sum_i \left| R(Q_i) - \hat{R}(Q_i) \right|^2\right]^{\frac{1}{2}} \quad (10)$$

$$l^1(\text{error}) = \sum_i \left| R(Q_i) - \hat{R}(Q_i) \right| \quad (11)$$

where $R(Q_i)$ is the measured reflectivity $I(Q_i) / I(0)$ and $\hat{R}(Q_i)$ is wavelet reconstruction of the reflectivity. Three wavelet families were used to analyze the scattered signal, D4, D10 and D28. The graphs are shown in Figs 1-3 and in Table I. The smoother, more regular D28 family performed better in the sense of the lower error norms at all noise removal levels. The widths in the last column of Table I are the average of all layers down to that layer. This contradicts the "Brownian motion" picture of a neutron in matter, but in the case of the GANS instrument, coherent effects dominate the specular scattering so this may not be surprising. Even though the numerical values in Table 1 contain several small disagreements from [2], the overall consistency of the trends are reassuring for both approaches. One counter-intuitive consequence [3], was that the Ti diffusion into NiC was much larger than the diffusion of NiC into Ti. This result was also found here in this very different analysis.

Table I. Results of the wavelet analysis of the neutron reflectivity measurements.

Peak	Q_M (nm^{-1})	σ_d^2 (nm^{-2})	$d(I,j) \pm \sigma_{d'}^{1}$ (nm)	$\sigma_{d'}^{1}$ (nm)
1	0.06539	0.0204		
2	0.12692	0.0215	5.025 ± 0.143	0.143
3	0.18942	0.0247	5.026 ± 0.146	0.146
4	0.25283	0.0216	4.955 ± 0.157	0.157
5	0.31247	0.2360	5.267 ± 0.147	0.147
6	0.37953	0.0148	4.686 ± 0.123	0.123

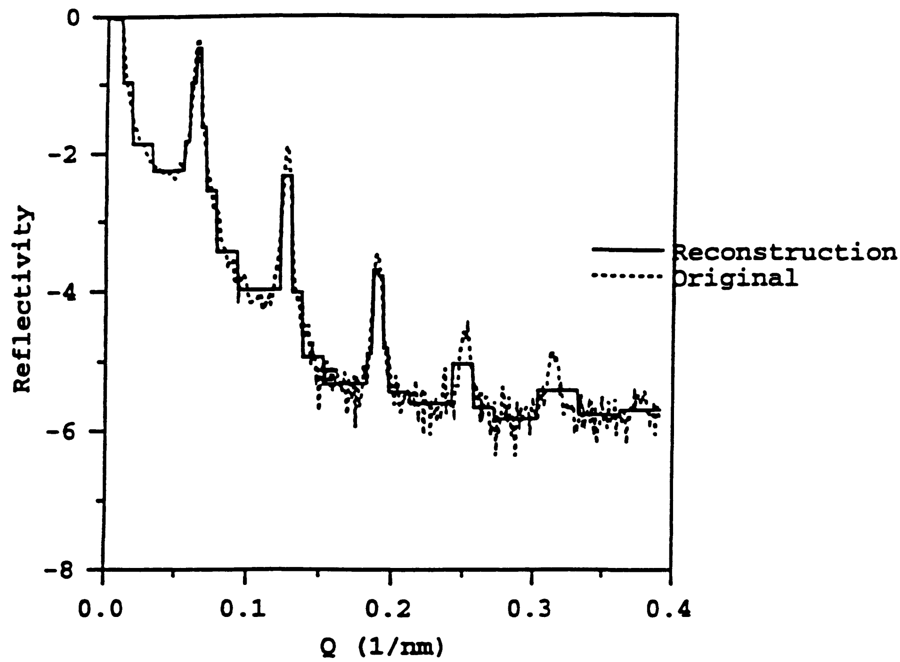

Figure 1. The measured neutron reflectance R (Q) vs wave number transferred Q in the dotted line is compared to the reconstruction using the rough D2 (Haar) wavelets in the solid line.

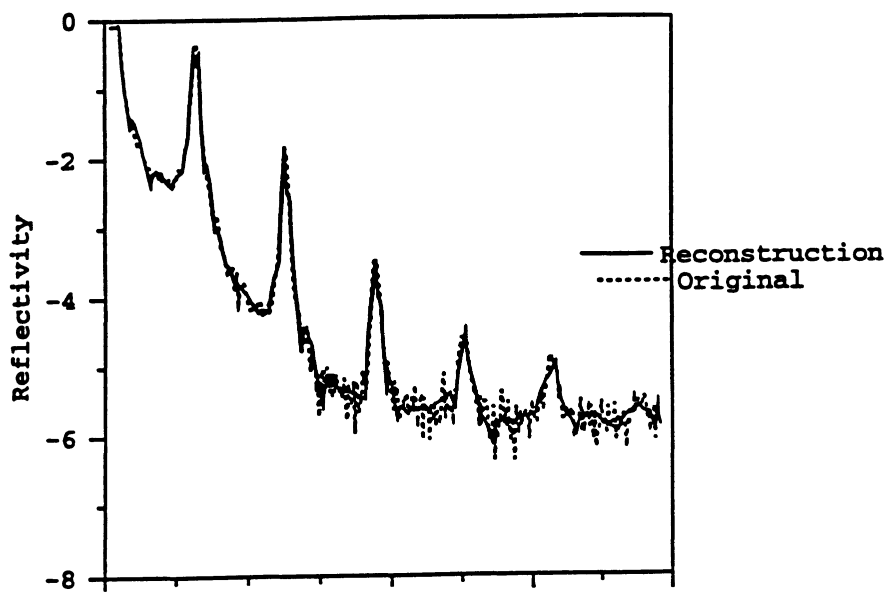

Figure 2. Plot of the measured R(Q) vs Q with the measured values in the dotted line and the reconstruction is shown in the solid line using the smoother D10 wavelets.

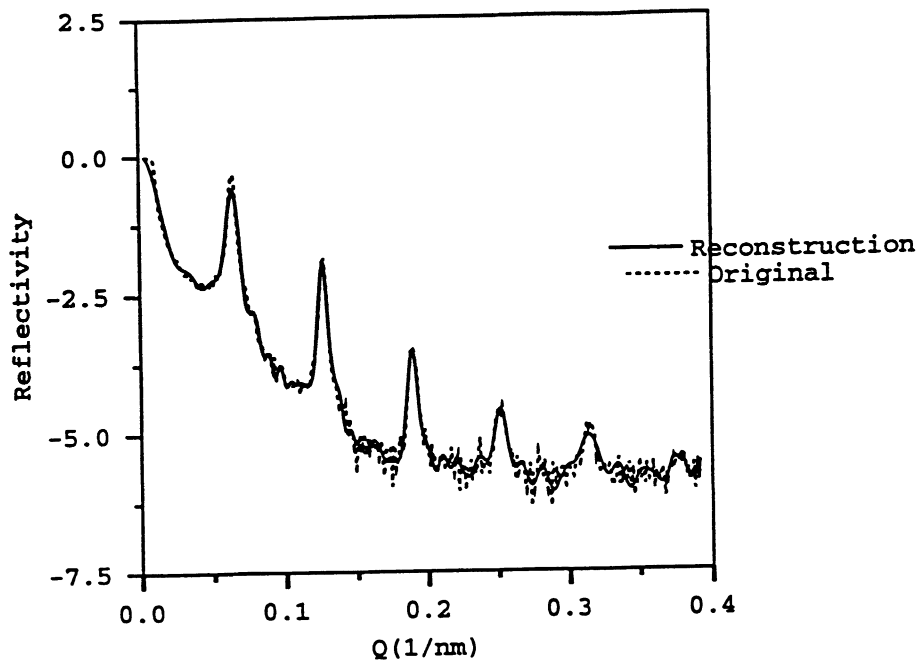

Figure 3. Plot of the measured R(Q) vs Q with the measured value shown in the dotted line and the D28 wavelet used in the solid line reconstruction.

It was shown that wavelet inverse methods applied to a metallic layered composite sample provide a variety of accurate information. This information was used to test the chemical vapor disposition, CVD, process quality of this composite device. Although, it is not possible to bring the reactor and the GANS spectrometer to the device, small composite structures can easily be brought to the reactor. Thus, NDE need not be restricted to bridges, pipelines and airplanes; and in the future it won't be.

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