



Foreword

I would like to begin by welcoming everyone to 8SXNS and thanking in particular Metin Tolan, Tim Salditt and Andreas Schreyer for organizing for us what is sure to be a very exciting conference. Since the second SXNS conference (and the first one that actually carried the name Surface X-ray and Neutron Scattering) was held in this place 13 years ago, the number of researchers working in this field has exploded. In addition, the field has matured enormously in the sophistication of the techniques used and the results obtained. Thus, it is worth taking a brief perspective of where we are currently in this area of science.

The most commonly used probes for investigating surfaces have been: low energy electron diffraction (LEED); auger spectroscopy; transmission electron microscopy (TEM); atomic beam scattering; scanning probe microscopies such as atomic force microscopy (AFM), scanning tunneling microscopy (STM), magnetic force microscopy (MFM), etc; probes using visible light, such as surface Raman scattering, second harmonic or sum difference frequency generation, and finally, the probes we are here to discuss, namely, neutron and X-ray scattering. These last probes usually involve large, expensive facilities, such as synchrotron X-ray sources or neutron sources. It is thus important to remind ourselves what is unique about scattering as a probe. Scattering, as opposed to imaging, is an *indirect* probe, which requires careful modeling of the data to obtain unique information, but it has the advantage that it is usually non-destructive, and provides us in one shot with global statistical information about the surface or interface and is thus ideal for studying collective behavior. It can also, unlike the scanning probe microscopies, probe buried interfaces or solid–liquid interfaces without too much of a problem, and can be used in a variety of environments, from low to high temperatures, with or without magnetic fields, and in various atmospheres at various pressures. Thus it is an important and unique probe. Nevertheless, it is also important to remember that it is not very useful to use scattering if the information sought can be obtained more easily (and cheaply) using some other technique.

Let us briefly remind ourselves how this field has developed. The first surface scattering studies using neutrons and X-rays were carried out (in the 1970s and 1980s) primarily on adsorbed monolayers and multilayers, often with randomly oriented or partially oriented high-surface-area substrates such as grafoil or intercalated graphite to overcome the intensity problems of seeing diffraction from a single surface [1]. These measurements were useful in providing unique information about 2D structure, order, disorder and collective phase transitions, such as commensurate–incommensurate and 2D melting transitions and 2D critical phenomena. They were complementary to absorption, heat capacity, LEED and atomic beam scattering experiments on these systems at that time. In addition, neutrons were used to probe the diffusional and vibrational dynamics of adsorbed molecules [2].

With the demonstration of the feasibility of grazing incidence X-ray diffraction (GIXD) from single surfaces [3], and the development of the concept and theory of truncation rod scattering [4], the stage was set for synchrotron X-rays to play a major role in carrying out what may be termed “surface crystallography”, to study effects such as surface reconstruction, faceting transitions, staging effects, etc. As

the scanning probe microscopies grew in popularity, they have tended to displace X-ray surface crystallography as the tool of choice for surface structure determination, but for *buried* interfaces, X-rays or neutrons are still the only probes that can be used without destroying the sample.

At the same time, the 1970s and 1980s witnessed a huge growth in the synthesis and study of artificially structured multilayer films. Reflectivity and diffraction from multilayers became commonly pursued studies at many laboratories with X-ray sources, as well as synchrotron sources to investigate issues such as roughness, interface diffusion, etc. of the multilayers. (At that time the importance of diffuse scattering was not properly appreciated, so many of these early studies did not bother to subtract the diffuse background.) The existence of multilayers was established via the observation of multilayer Bragg peaks in the specular reflectivity. (We use the term here to apply specifically in the small Q region). Already, in the 1950s, Parratt [5] had developed his famous iterative scheme for analyzing specular reflectivity from a stack of different layers, generalizing methods used in optics [6], and this provided an in-principle rigorous method, which is used, with certain modifications to this day!! Nevot and Croce [7] came along and showed how the Fresnel reflectivity formula could be modified to take into account surface roughness, and soon this was incorporated ad hoc also into the Parratt formalism.

While the spin dependence of neutron reflectivity from a magnetic surface had been known from early days, actual studies of magnetism in thin films using the magnetic specular reflectivity of polarized neutrons was pioneered by Felcher and coworkers [8] at the relatively low-intensity pulsed neutron source at Argonne. This has now become a very popular type of experiment around the globe today which has grown enormously in sophistication of both technique and analysis. Around the same time, Als-Nielsen, Pershan and their collaborators pioneered the use of X-ray specular reflectivity to study liquid surfaces [9]. Other uses of neutron and X-ray specular reflectivity (as opposed to grazing incidence diffraction), which developed rapidly during the 1980s and 1990s were the study of polymer films, surfactants and microemulsions, Langmuir–Blodgett multilayers, and even biological systems such as protein layers [10]. The technique was also used to study wetting phenomena [11], surface melting [12] or surface freezing [13]. With the discovery of resonant magnetic X-ray scattering, X-ray specular reflectivity in the mid 1990s also began to be used to complement neutron reflectivity to study magnetization depth profiles in thin films and multilayers in an element-selective manner, with soft X-rays being used to study transition metals and hard X-rays for rare-earth metals [14].

In the late 1980s and 1990s attention also began to focus on the information contained in off-specular scattering, particularly from rough and disordered surfaces and interfaces. Detailed treatments of this difficult problem in the context of light scattering from rough surfaces had already been published earlier, but the treatments were complicated. A simpler treatment using the Born approximation and the distorted wave Born approximation (DWBA) [15] and a Gaussian variable assumption for the roughness fluctuations enabled the diffuse scattering for X-rays and neutrons to be expressed (and fitted) in terms of height–height correlation functions. Then a simple self-affine model allowed surface roughness to be parametrized in terms of only 3 parameters. Soon these methods were generalized to multiple surfaces and multilayers [16], laterally structured surfaces [17], liquid surfaces [18], and most recently magnetic surfaces [19]. Off-specular surface scattering at grazing incidence has been used to study the morphology of surface and interface roughness, wetting films, film growth exponents, capillary waves on liquid surfaces, magnetic roughness and the morphology of magnetic domains in magnetic films.

Over the last decade, grazing incidence diffraction with X-rays and more recently also with neutrons has been used to study arrays of nanodots, magnetic hole arrays, etc. Specular and off-specular measurements and GIXD (particularly at high photon energies) have been used to probe the ordering of liquids and the growth of crystals at solid–liquid interfaces, surface electrochemistry, liquid–liquid interfaces and fluids confined between solid surfaces.

The equations governing specular reflectivity are known to exhibit certain guided wave resonances under certain conditions [20], which have been exploited to make X-ray and neutron waveguides [21] and to

enhance the intensity for grazing incidence diffraction from thin films [22]. Finally, the use of coherent X-ray beams available from the new synchrotron sources has also been applied to surface scattering to observe surface speckle (and even magnetic speckle in the case of magnetic films), which in turn has been used for image reconstruction of surfaces [23], and in the form of X-ray photon correlation spectroscopy at grazing incidence [24] to study slow dynamical surface fluctuations. The latter technique can be complemented by grazing incidence neutron spin echo experiments, as demonstrated by de Jeu and coworkers at this meeting.

Finally, let me turn to some issues in our field which are still in the process of being resolved. These include questions such as: Can we use diffuse scattering to identify details of roughness at the atomic level, which might take the form of pyramids, cylinders, steps, islands or bubbles? How does one distinguish between “roughness” vs. “flatness”? How does one distinguish broadening due to surface mosaic from that due to diffuse scattering? How does roughness (structural and magnetic) affect transport and magnetotransport in thin films, or the coercive field or the magnetic anisotropy? How is it related to domain formation? What information does speckle ultimately carry? Can one invert speckle patterns to image magnetic domains? Can one use grazing incidence diffraction to verify in-plane ordering in fluids confined at the spacing of a few molecular diameters? What are the dynamical fluctuations in confined as opposed to bulk fluids?

My attempt to do some crystal-ball gazing in connection with future developments in this field came up with the following areas, which are likely to grow in importance: surface or thin film inelastic scattering with both neutrons and X-rays; XPCS coupled with grazing incidence NSE; imaging with coherent beams; biological systems; the study of nanostructures; pump–probe methods combined with reflectivity to study effects due to shock waves, pulsed magnetic fields, etc.: and surface magnetism and phase transitions. Quite probably, I will be proved wrong in some instances, but I think it *is* safe to say that this field will stay vigorous and exciting for several more SXNS conferences!!

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