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2.9. Neutron reflectometry

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2.9.1. Introduction

The neutron reflectivity of a surface is defined as the ratio of the number of neutrons elastically and specularly reflected to the number of incident neutrons. When measured as a function of neutron wave-vector transfer, the reflectivity curve contains information regarding the profile of the in-plane average of the scattering-length density (or simply scattering density) normal to that surface. The concentration of a given atomic species at a particular depth can then be inferred. Although similar information can often be extracted from X-ray reflectivity data, an additional sensitivity is gained by using neutrons in those cases where elements with nearly the same number of electrons or different isotopes (especially hydrogen and deuterium) need to be distinguished. Furthermore, if the incident neutron beam is polarized and the resultant polarization of the reflected beam analysed, it is possible to determine, in both magnitude and direction, the in-plane magnetic moment depth profile. This latter capability is greatly facilitated by the simple correlation of the relative counts of neutron spin-flip and non-spin-flip scattering to magnetic moment orientation and by the relatively high instrumental polarization and spin-flipping efficiencies possible with neutrons. These properties make neutron reflectivity a powerful tool for the study of surface layers and interfaces.

2.9.2. Theory of elastic specular neutron reflection

Consider the glancing (small-angle) reflection of a neutron plane wave characterized by a wave vector $\mathbf{k}_i$ from a perfectly flat and smooth surface of infinite lateral extent, as depicted schematically in Fig. 2.9.2.1. Although the density of the material can, in general, vary as a function of depth [along the direction (z) of the surface normal], it is assumed that there are no in-plane variations of the density. If the scattering is also elastic, so that the neutron neither gains nor loses energy (*i.e.* $|\mathbf{k}_i| = |\mathbf{k}_f| = k = 2\pi/\lambda$, where the subscripts i and f signify initial and final values, respectively, and λ is the neutron wavelength), then the component of the neutron wave vector parallel to the surface must be conserved. In this case, the magnitude of the wave-vector transfer is $Q = |\mathbf{Q}| = |\mathbf{k}_f - \mathbf{k}_i| = 2k\sin(\theta) = 2k_z$, where the angles of incidence and reflection, θ, are equal, and the scattering is said to be *specular*. Since at low values of Q the neutrons are strongly scattered from the surface (*i.e.* the magnitude of the reflectivity approaches 1), the neutron wave function is significantly distorted from its free-space plane-wave form. The first Born approximation normally applied in the description of high-Q crystal diffraction is therefore not valid for the analysis of low-Q reflectivity measurements, and a more accurate, dynamical treatment is required.

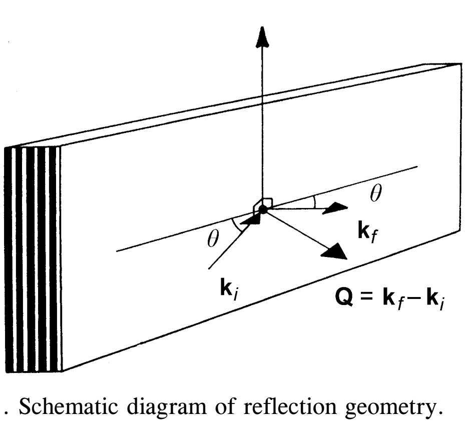

Fig. 2.9.2.1. Schematic diagram of reflection geometry.

Because the in-plane component of the neutron wave vector is a constant of the motion in the specular elastic reflection process described above, the appropriate equation of motion is the one-dimensional, time-independent, Schrödinger equation (see, for example, Merzbacher, 1970)

$$\psi''(z) + k_z^2\psi(z) = 0, \qquad (2.9.2.1)$$

where ψ is the neutron wave function [which in free space is proportional to $\exp(ik_{0z}z)$, where k_{0z} is the magnitude of the z component of the neutron wave vector in vacuum]. If the infinite planar boundary from which the neutron wave reflects separates vacuum from a medium in which the neutron potential energy is V_1, conservation of the neutron's total energy requires that

$$k_{0z}^2 = k_{1z}^2 + \frac{2mV_1}{\hbar^2}, \qquad (2.9.2.2)$$

where $\hbar$ is Planck's constant divided by 2π and m is the neutron mass. If $2\pi/Q$ has a magnitude much greater than interatomic distances in the medium, then the medium can be treated as if it were a continuum. In this limit, the potential energy V_1 can be expressed as (see, for example, Sears, 1989)

$$V_1 = \frac{2\pi\hbar^2\overline{Nb}}{m}, \qquad (2.9.2.3)$$

where $\overline{Nb} = \sum N_i b_i$, i represents the ith atomic species in the material, N_i is the number density of that species and b_i is the coherent neutron scattering length for the ith atom (which is in general complex if absorption or an effective absorption such as isotopic incoherent scattering exists; magnetic contributions are not accounted for here but will be considered below). The quantity $\overline{Nb} \equiv \rho$ is the effective scattering density. Substituting the expression for V_1 given in equation (2.9.2.3) into (2.9.2.2) yields

$$k_{1z}^2 = k_{0z}^2 - 4\pi\rho(z). \qquad (2.9.2.4)$$

In order to calculate the reflectivity, continuity of the wave function and its first derivative (with respect to z) are imposed. These boundary conditions are a consequence of restrictions on current densities required by particle and momentum conservation. In general, given a sample with layers of varying potentials where the boundaries of the jth layer are at z_{oj} and $z_{oj} + \delta_j$, and the potential, V_j, is constant over that layer, it can be shown that (see, for example, Yamada, Ebisawa, Achiwa, Akiyoshi & Okamoto, 1978)

$$\begin{pmatrix}\psi_j(z_{oj}+\delta_j)\\ \psi'_j(z_{oj}+\delta_j)\end{pmatrix} = \mathbf{M}_j\begin{pmatrix}\psi_j(z_{oj})\\ \psi'_j(z_{oj})\end{pmatrix} = \begin{pmatrix}\psi_{j+1}(z_{oj}+\delta_j)\\ \psi'_{j+1}(z_{oj}+\delta_j)\end{pmatrix}, \qquad (2.9.2.5)$$

where

$$\mathbf{M}_j = \begin{pmatrix}\cos(k_{jz}\delta_j) & (1/k_{jz})\sin(k_{jz}\delta_j)\\ -k_{jz}\sin(k_{jz}\delta_j) & \cos(k_{jz}\delta_j)\end{pmatrix}, \qquad (2.9.2.6)$$

references