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Retrieval of the reflection coefficient in spin-polarized neutron specular reflectometry

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Abstract

A method is proposed which allows a complete determination of the complex reflection coefficient for any free unknown real potential (i.e., in the case where there is no effective absorption). It exploits the interference of the spin components of a polarized neutron beam in the presence of a magnetic reference layer mediated between substrate and sample. Following the recent investigations, the method requires polarization analysis of the reflected beam instead of reflectivity analysis. However, our method is based on the transfer matrix formalism. In this method we use the relations between the polarization and the reflectivity of the reflected beam and the transfer matrix elements for the known and unknown samples. Using these relations we show that instead of fully polarization measurement, some suitable choice of polarization and reflectivity measurements can be used to determine the reflection coefficient. The method is supplemented with a schematic example.

1. Introduction

Specular reflectivity of cold and ultracold neutrons can provide important information about physical and chemical phenomena occurring at surfaces and interfaces of condensed matter [1–4]. Hence, the use of such neutron reflection to study thin films and interfaces has increased dramatically. In neutron reflection, the interaction of neutrons with the film's atomic constituents is characterized by a continuous scattering length density (SLD) function which is a number-density-weighted microscopic average of known isotope-specific constants, the scattering lengths. So the SLD can determine the coherent elastic scattering behaviour of neutrons by the microstructure of a film [5].

The major aim of neutron specular reflectometry is to reveal the in-plane average of the SLD depth profile in the direction normal to the surface of materials in thin film geometries [2]. The SLD depth profile of a sample can be directly converted to the chemical profile of the sample [6].

The SLD depth profile determines the specular reflection from a film. So it can be obtained from measurements of the reflectivity, R(q). The reflectivity is defined as the number of

neutrons reflected elastically and specularly as a function of the glancing angle of incidence reflection, relative to the plane of the film, or as a function of the normal component of the incident neutron wavevector, q.

However, extracting the profile from measured reflectivity has been hampered by the socalled *phase problem* [7–9]. This problem, widely discussed in structure analysis, refers to the fact that in reflection experiments only the square of the complex reflection coefficient, $R(q) = |r(q)|^2$, is measured, and as in any other scattering technique, the phase of the reflection is lost [10]. In the absence of the phase, least-squares methods [11, 12] are generally used to extract the SLD profile, but in general more than one SLD may be found to correspond to the same reflectivity [13]. A simple example is the difference between a freestanding SLD profile, $\rho(x)$, and its mirror image, $\rho(L - x)$, where *L* is the thickness of the film and *x* is the depth from the surface. Both of these quantities produce the same R(q) with different r(q)-values; only the phase of reflection distinguishes one from the other. So, given the phase, it is possible to solve the one-dimensional inverse scattering problem directly to obtain a unique SLD depth profile [14–17]. Thus, at first, it is necessary to have a reliable and practical way of determining the reflection coefficient from reflectivity measurements.

In general, the reflection phase is not entirely independent of the reflectivity, owing to the analytic properties of the complex reflection coefficient (there exists a dispersion relation between the logarithm of the reflectivity and the reflection phase [18]). For a real potential which vanishes on the negative half-axis, V(x) = 0 for x < 0, and supports no bound states, which corresponds to most actual situations in neutron reflectometry, the complex reflection coefficient can be written as $r(q) = A(q)r_H(q)$. The 'Hilbert reflection' coefficient, $r_H(q)$, is completely determined by the reflectivity R(q), whereas the rational phase factor A(q)involving the zeros of the reflection coefficient in the upper half-plane is unknown in the absence of phase data. Thus, the phase problem reduces to the problem of finding the zeros of the reflection coefficient in the upper half-plane, which represent the remaining ambiguity in solving the inverse scattering problem with the reflectivity as input [7, 19].

Apart from these mathematical considerations, several methods for actual phase determination have been developed, such as the reference layer method [20–31] and the dwell time method [32]. In the former method, a known reference layer having tunable values of the SLD is used to exactly determine the reflection amplitude or the phase. The latter method makes use of a relation between the dwell time and the phase times [33]. This relation is transformed into a differential equation for the reflection phase as a function of neutron wavenumber containing as known functions the reflectivity and dwell time.

Among these methods, the reference layer method (see [20] for a review) seems the best, because of its application in experiment. The experimental implementation of the reference layer method was first achieved with good success by Majkrzak *et al* [22], who also proposed and tested experimentally the related surrounding method [23, 24].

The reference layer methods based on polarization measurements [28-31] are of particular interest, since they also work in the total reflection regime and allow unique reconstruction of surface profiles of magnetic samples. The use of magnetic reference layers has this extra advantage that the state of the layer can be changed without a chemical or structural effect on the sample. In this method r(q) can be determined by measuring the reflectivity or polarization of the reflected neutrons. If the reflectivity is measured, it is not important whether the reference magnetic layer is mounted on the top or bottom of the unknown layer. This is due to the fact that for the reflection coefficient of the reversed unknown layer) can be determined. However, for the reflected neutron the bottom, the polarization analysis method proposed in [29] cannot be used, because this method uses the fact that the wavefunction for scattering 'from the left' by a sample on a substrate of finite thickness can be expressed in terms of the corresponding 'left' and 'right' scattering functions for a bulk substrate [34]. In this case, Leeb *et al* [30] have shown that the reflection from the right-hand side of the unknown absorptive non-magnetic sample can be determined by at least two polarization measurements with different reference layers.

In this paper, we propose an efficient method based on the polarization analysis for determining the reflection coefficient of an unknown non-absorptive layer mounted on top of the magnetic reference layer. The method is similar to the one proposed in [29] in that in both of them one needs polarization measurements. However, our mechanism is based on the transfer matrix theory. The description within the formalism of the transfer matrix for non-vacuum surrounding media has been investigated in [35] theoretically. In that paper, the refractive indices of the substrate and the media in front enter into our calculations through the transfer matrix. Also, there, the reflection coefficient of the reversed layer surrounded on both sides by the same fronting media has been obtained. Here, we investigate this method for vacuum fronting media; hence the reflection coefficient of the reversed free unknown layer can be extracted. Finally we illustrate our proposed method by a specific example.

The layout of the paper is as follow. In section 2, we obtain the basic relations between the polarization components of the reflected beam as a function of the elements of the transfer matrix of the unknown and known layers. Using these relations, we show how the reflection coefficient of the reversed unknown layer can be extracted by fully polarization analysis. However, it is shown that if we are provided with the knowledge of the reflectivity measurements, then there is no need to have complete polarization measurements. In this case, some suitable measurements of polarization and reflectivity of the reflected beam is sufficient. In section 3, a schematic example is presented as an illustration of our method. The paper is ended by a conclusion.

2. The method

Here we are going to explain our method. Consider that the sample is mounted on top of a thick substrate having constant SLD, ρ_s . The sample is separated into two distinct films, with the unknown layer mounted on the magnetic reference layer. The reference layer is magnetized in a direction parallel to the reflecting surface which is taken as the direction of spin quantization. It is assumed that the magnetic field vanishes in the unknown sample and the incident beam is polarized in the direction normal to the reflection surface. In this case, the polarization of the reflected beam can be expressed as follows:

$$P_z = \frac{R_+(q) - R_-(q)}{R_+(q) + R_-(q)},\tag{1}$$

$$P_x + iP_y = \frac{2r_+^*(q)r_-(q)}{R_+(q) + R_-(q)},$$
(2)

where $r_+(q)$ and $r_-(q)$ are the reflection coefficients for the neutron beam polarized, respectively, parallel and antiparallel to the magnetic field, and $R_{\pm} = |r_{\pm}|^2$.

 $r_{\pm}(q)$ can be derived from the transfer matrix method of solving a one-dimensional Schrödinger equation,

$$\partial_x^2 \Psi(q, x) + 4\pi \rho(x) \Psi(q, x) = q^2 \Psi(q, x).$$
(3)

The 2 × 2 unimodular transfer matrix has elements A(q), B(q), C(q) and D(q) that are completely determined by knowing the SLD depth profile of the sample, $\rho(x)$. Here we must emphasize an important assumption. Henceforth we take advantage of the fact that

the SLD of neutrons almost always can be taken to be real; the imaginary part accounts for absorption, which is negligible for neutrons, and for thin films, even when incoherent scatterers (such as water) are involved [1, 5]. As a result of this assumption, the transfer matrix elements are real-valued at all q. In terms of these elements, the solution of equation (3) for vacuum fronting media leads to [20]

$$r(q) = \frac{(h^2 B^2 + D^2) - (h^2 A^2 + C^2) - 2i(h^2 A B + CD)}{(h^2 B^2 + D^2) - (h^2 A^2 + C^2) + 2h},$$
(4)

where *h* is determined by ρ_s as below:

$$h = (1 - 4\pi\rho_{\rm s}/q^2)^{1/2}.$$
(5)

The reflectivity, R_{\pm} , can be related to the elements of the transfer matrix in terms of the new quantity $\Sigma_{\pm}(q)$:

$$\Sigma_{\pm} = 2\frac{1+R_{\pm}}{1-R_{\pm}} = h(A^2 + B^2) + \frac{1}{h}(C^2 + D^2).$$
(6)

By introducing the following new parameters, as

$$\alpha^{h} = hA^{2} + h^{-1}C^{2}, \tag{7a}$$

$$\beta^n = hB^2 + h^{-1}D^2, \tag{7b}$$

$$\gamma^h = hAB + h^{-1}CD, \tag{7c}$$

equations (4) and (6) can be defined in the form

$$r(q) = \frac{\beta^h - \alpha^h - 2i\gamma^h}{\alpha^h + \beta^h + 2},\tag{8}$$

$$\Sigma_{\pm} = \alpha^h + \beta^h, \tag{9}$$

in which 'h' as superscript denotes that the sample is mounted on top of a substrate having refractive index h. These two equations show that the complex reflection coefficient is determined locally by three real parameters α , β and γ , while the reflectivity depends only on $\alpha + \beta$. Thus, in general, R(q) cannot determine r(q) locally in q. This is essentially an alternative restatement of the phase problem.

As in our sample the SLD profile is separated into two distinct regions representing the known magnetic and the unknown parts, the total transfer matrix can be expressed as a product of the corresponding transfer matrices:

$$\begin{pmatrix} A & B \\ C & D \end{pmatrix} = \begin{pmatrix} w_{\pm} & x_{\pm} \\ y_{\pm} & z_{\pm} \end{pmatrix} \begin{pmatrix} a & b \\ c & d \end{pmatrix},$$
(10)

where the matrix (a, \ldots, d) describes the contribution from the unknown part of ρ and $(w_{\pm}, \ldots, z_{\pm})$ gives the known part. The signs, (+) and (-), denote the plus and minus magnetization of the reference layer with respect to the neutron beam polarized parallel and antiparallel to the magnetic field.

In this case, using equations (7) and (10), we can write

$$\alpha_{+}^{h} = \alpha_{k+}^{h} a^{2} + \beta_{k+}^{h} c^{2} + 2\gamma_{k+}^{h} ac, \qquad (11a)$$

$$\beta_{+}^{h} = \alpha_{k+}^{h} b^{2} + \beta_{k+}^{h} d^{2} + 2\gamma_{k+}^{h} bd, \qquad (11b)$$

$$\gamma^h_{\pm} = \alpha^h_{k\pm}ab + \beta^h_{k\pm}cd + 2\gamma^h_{k\pm}(ad+bc), \qquad (11c)$$

where α_{\pm}^{h} , β_{\pm}^{h} and γ_{\pm}^{h} denote the entire arrangement containing the known film, magnetic reference layer, and the substrate, and $\alpha_{k\pm}^{h}$, $\beta_{k\pm}^{h}$ and $\gamma_{k\pm}^{h}$ denote the magnetic reference layer mounted on top of the substrate (i.e., the entire arrangement without the unknown sample).

Using equations (11*a*) and (11*b*), we can write

$$\Sigma_{\pm}(q) = \beta_{k\pm}^{h} \tilde{\alpha}_{u} + \alpha_{k\pm}^{h} \tilde{\beta}_{u} + 2\gamma_{k\pm}^{h} \tilde{\gamma}_{u}, \qquad (12)$$

where the tilde denotes a reversed unknown film; that is, the interchange of the diagonal elements of the corresponding transfer matrix $(a \leftrightarrow d)$ [36]. Using equation (8) it is obvious that given the parameters $\tilde{\alpha}_u$, $\tilde{\beta}_u$, and $\tilde{\gamma}_u$, the complex reflection coefficient of the reversed free unknown film can be determined.

To show the ability of finding these unknown parameters by polarization measurements, we first derive the polarization of the reflected beam as a function of the transfer matrix elements of the known and unknown layers. Using equations (1), (2), (8), (9), and (11), after some straightforward (yet tedious) algebra, we obtain

$$P_x = 1 + \frac{2\zeta}{\Sigma_+ \Sigma_- - 4},\tag{13}$$

$$P_{y} = \frac{2}{\Sigma_{+}\Sigma_{-} - 4} (c_{\gamma\beta}\tilde{\alpha}_{u} + c_{\alpha\gamma}\tilde{\beta}_{u} + c_{\alpha\beta}\tilde{\gamma}_{u}), \qquad (14)$$

$$P_z = 2\frac{\Sigma_+ - \Sigma_-}{\Sigma_+ \Sigma_- - 4},\tag{15}$$

where

$$\zeta = 2(1 + \gamma_{k+}^{h} \gamma_{k-}^{h}) - (\alpha_{k+}^{h} \beta_{k-}^{h} + \beta_{k+}^{h} \alpha_{k-}^{h}),$$
(16)

and

$$c_{ij} = i^h_{k+} j^h_{k-} - j^h_{k+} i^h_{k-} \tag{17}$$

for *i* and $j = \alpha', \beta'$, and γ' . Equations (16) and (17) show that ζ and c_{ij} are independent of the unknown layer and are determined by the transfer matrix elements of the reference layer and the refractive index of the substrate. For non-vacuum fronting media, these parameters depend on the refractive index of the fronting media too [35].

Equations (13)–(15) are essential in our method. Dependence on unknown parameters in P_x and P_z is encoded in the parameters Σ_+ and Σ_- . However, P_y , aside from Σ_{\pm} (in the denominator), depends on the unknown sample by a linear combination of the unknown parameters in the numerator. It also should be emphasized that such relations, as equations (13)–(15), are not valid when the unknown layer is mediated between the reference layer and the substrate. It is also worth nothing that in equations (13)–(15) we could use the transfer matrix elements of the known layer surrounded at both sides by a uniform layer having constant SLD of ρ_h ; $\alpha_{k\pm}^{hh}$, $\beta_{k\pm}^{hh}$ and $\gamma_{k\pm}^{hh}$. It is due to the fact that $i^h = h \times i^{hh}$ for $i = `\alpha'$ and ` β ', and $\gamma^h = \gamma^{hh}$.

As is seen from equations (13) and (15), when we are provided with fully polarization measurements, Σ_+ and Σ_- can be obtained as follows:

$$\Sigma_{\pm}^{2} \mp \frac{\zeta P_{z}}{2(P_{x} - 1)} \Sigma_{\pm} - \left(4 + \frac{\zeta}{P_{x} - 1}\right) = 0.$$
(18)

This quadratic equation has two different solutions; the physical solution must be selected using the fact that $\Sigma_{\pm} > 2$. Although numerical examples show that only one root satisfies $\Sigma_{\pm} > 2$, we have no general proof of this. This is similar to the situation in Leeb *et al* [31].

Once Σ_+ and Σ_- are determined, we have two linear relations for the unknown parameters, equation (12). Another relation can be determined by the measurement of P_y . Aside from fully polarization measurements, equations (13)–(15) show that Σ_+ and Σ_- and their corresponding linear relations (equation (12)) can be obtained by more flexible ways. However, these ways are applicable if we have the knowledge of the reflectivity measurements, i.e., the knowledge

of Σ_+ or Σ_- . Against the fully polarization measurement, in this case the polarization of the incident beam should be changed.

One of the advantages of these alternative ways is that here the problem of selecting the physical solution is not faced. As is seen from equations (13) to (15), by knowing two of the four parameters, Σ_+ , Σ_- , P_x , and P_z , the others can be found. Thus, if we are provided with one of the reflectivity measurements, i.e., either Σ_+ or Σ_- , the other can be obtained by measuring P_x and P_z . Therefore, Σ_+ and Σ_- can be obtained by measuring two of the four parameters, Σ_+ , Σ_- , P_x and P_z . The critical point is that measurement of the polarization of the reflected beam parallel to the sample surface and normal to the magnetization direction of the reference layer is necessary in all cases. It should be mentioned that for an incident beam fully polarized in the y-direction, measurement of P_x is necessary.

For example, when we measure the reflectivity for an incident neutron beam fully polarized in the direction antiparallel to the magnetic field, and P_x and P_y for an incident neutron beam fully polarized normal to the surface of the sample, the unknown parameters can be calculated as follows:

$$\begin{pmatrix} \tilde{\alpha}_u \\ \tilde{\beta}_u \\ \tilde{\gamma}_u \end{pmatrix} = M^{-1} \begin{pmatrix} \Sigma_- \\ \frac{1}{\Sigma_-} \left(4 + \frac{2\zeta}{P_x - 1} \right) \\ \zeta P_y / (P_x - 1) \end{pmatrix},$$
(19)

where

$$M = \begin{pmatrix} \beta_{k-}^h & \alpha_{k-}^h & 2\gamma_{k-}^h \\ \beta_{k+}^h & \alpha_{k+}^h & 2\gamma_{k+}^h \\ c_{\beta\gamma} & c_{\alpha\gamma} & c_{\alpha\beta} \end{pmatrix}.$$
 (20)

Combination of the polarization and reflectivity measurements of the reflected beam cannot be used by the method of [29], since in that method knowledge of the reflectivity measurements yields only an alternative way to select the physical coefficient, i.e., in any case the fully polarization measurements is needed.

3. Example

As a realistic example to test the method by simulation, we consider the arrangement of Leeb *et al* [34] with a magnetic reference layer, an unknown sample, and a substrate (e.g. a Si wafer), without Cd film because in our method there is no absorption.

The sample is a 30 nm thick gold layer on a 20 nm Cr layer having constant SLD values of 4.46×10^{-4} nm⁻² and 3.03×10^{-4} nm⁻², respectively. The magnetic reference layer is a 15 nm thick Co layer having SLD values of 6.44×10^{-4} , -1.98×10^{-4} and 2.23×10^{-4} nm⁻², respectively for plus, minus and non-magnetization. The reference layer is magnetized in a direction parallel to the reflecting surface (+*z*-direction) which is taken as the direction of the spin quantization. Since an external field is necessary to magnetize the Co film in the desired direction, it may affect the neutron beam. It is assumed that the associated magnetic induction outside this film is small enough not to affect the neutron beam appreciably (in particular, its spin); we set it equal to zero for simplicity. The roughness of interfaces is neglected.

Figure 2 shows the q-dependence of the polarization of the reflected beam $(P_x, P_y, \text{ and } P_z)$, when the incident beam is fully polarized in the +x-direction.

Figure 3 shows the reflectivity of the reflected beam, R_{\pm} , when the incident beam is fully polarized in the $\pm z$ -direction. In all calculations the simulated data start at the critical q of the substrate.



Figure 1. Experimental arrangement for measuring the complex reflection coefficient. Top: arrangement of the layers. Bottom: the SLD depth profile. The dotted lines represent the effective SLD experienced by neutron beams polarized parallel and antiparallel to the magnetic field **B**.



Figure 2. Simulated polarization data of the reflected beam for the entire arrangement shown in figure 1. The incident beam is assumed to be fully polarized in +x-direction. The simulated data start at the critical q of the substrate (P_x : dotted curve, P_y : dashed curve, P_z : solid curve).

Figure 4 shows the real and imaginary parts of the reflection coefficient for the mirror image of the unknown sample (i.e. the Cr layer is mounted on top of the gold layer) derived from the data of P_x and P_y (in figure 2), and R_- (in figure 3) and using equation (17).

Below the critical q_c , $q_c^2 = 4\pi\rho_s$, for total external reflection by the substrate, the facts that Re $r(q) \rightarrow -1$ and Im $r(q) \rightarrow 0$ make it possible to reliably interpolate in this region using a slab potential of adjustable width and height, determined by fitting the measured reflectivity near q_c [22]. In addition, since we have neglected any absorption, we have $r(-q) = r^*(q)$, so that the reflection coefficient of the reversed free unknown film is obtained for the entire range of q. Once $\tilde{r}(q)$ has been determined for the whole range of q, it can be converted to the desired $\rho(L-x)$ by solving the Gel'fand–Levitan–Marchenko integral equations [16, 17, 21], where L is the thickness of unknown sample.



Figure 3. Simulated reflectivity data for the entire arrangement shown in figure 1. The reflectivity of the reflected beam when the incident beam is fully polarized in the +z and -z directions are shown by solid and dashed curves, respectively.



Figure 4. Real and imaginary part of the reversed free unknown film of figure 1 (i.e. the Cr film is mounted on top the gold film). Solid curve: computed directly from equation (4). Circles: recovered from the data of figures 2 and 3.

As figure 4 shows, the recovered data by using the data of $(P_x, P_y \text{ and } R_-)$ are perfect. This is reasonable because the exact data are used. To test the stability of the method in reconstruction of the complex reflection coefficient in the presence of experimental uncertainties, we have simulated measurement errors by using input ensembles of normally distributed polarization values with half-width = 0.02. Similarly, the reflectivities R_- are taken as normal distributions with half-width = $0.05R_-$. Also, to include surface roughness, we have randomly varied the position of the interfaces within a width of 0.5 nm around the mean value. These uncertainties are similar to the ones used by Kasper *et al* [31]. So, to compare the stability of our method against the method of [31], we reconstruct the amplitude and the phase



Figure 5. Reconstruction of the amplitude and the phase of the complex reflection coefficient extracted from simulated P_x , P_y and R_- for the arrangement shown in figure 1. The simulation includes the effect of surface roughness and assumes mean uncertainties of 0.02 and 0.05 R_- of the polarization and reflectivity, respectively.

of the complex reflection coefficient, r, instead of its real and imaginary part. By using this statistical ensemble of data as input, and applying equation (19), we reconstruct the amplitude and the phase of r as displayed in figure 5, where the exact reflection coefficient is shown too. This figure shows a isolated region near the critical wavenumber where the solution is rather unstable. These large uncertainties in the vicinity of the critical wavenumber are corroborated by the fact that in the presence of statistical errors, the condition of equation (19) becomes ineffective. However, except for this isolated region, the results are in fair agreement with the exact values.

4. Conclusions

Without knowledge of the phase of the reflection coefficient or without sufficient additional physical information, a single set of reflectivity data for a non-symmetric sample film alone can only reveal whether a particular SLD profile is consistent with the data, but this is not the only possible result. The phase of the reflection coefficient can be used as an additional piece of information to guarantee the uniqueness of an SLD profile, so knowing the complex reflection coefficient r(q) is necessary.

In this paper we have proposed a method to determine the complex reflection coefficient of an unknown non-absorptive layer by using a magnetic reference layer mediated between the unknown layer and the substrate and polarization analysis of the reflected neutrons in the formalism of the transfer matrix. The method is based upon relations between the polarization of the reflected beam and transfer matrix elements of the known and unknown layers. Since the substrate is included in the formalism by the refractive index h, the known parameters do not change by changing the substrate. Also, we show that given the results of reflectivity measurements of the reflected beam, more flexible ways can be used to extract the complex reflection coefficient. However, in these cases, it has been shown that for an incident beam fully polarized normal to the sample surface the measurement of P_y is essential.

Also, in our analysis we have treated experimental uncertainties. It has been shown that they do not affect the determination of the reflection coefficient, except around the critical wavenumber.

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