Retrieval of phase information in neutron reflectometry

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Neutron reflectometry can determine unambiguously the chemical depth profile of a thin film if both phase and amplitude of the reflectance are known. The recovery of the phase information is achieved by adding to the unknown layered structure a known ferromagnetic layer. The ferromagnetic layer is magnetized by an external magnetic field in a direction lying in the plane of the layer and subsequently perpendicular to it. The neutrons are polarized either parallel or opposite to the magnetic field. In this way three measurements can be made, with different (and known) scattering-length densities of the ferromagnetic layer. The reflectivity obtained from each measurement can be represented by a circle in the (complex) reflectance plane. The intersections of these circles provide the reflectance.

Neutron reflection experiments are important in understanding the physics of many surface and interfacial structures, in fields as diverse as polymer science[1] and magnetism.[2] The measurement of the reflectivity profile $R(q)$, where $q=2\pi\sin\theta/\lambda$, in terms of the neutron wavelength λ and the reflection angle θ, gives information about the atomic or magnetic density profile of the samples along its depth z. The reflectivity is the square of the amplitude of the reflectance, $r(q)$. If the reflectance is known in amplitude and phase, the transformation

$$r(q)\leftrightarrow\Gamma(z)=4\pi b(z)N(z) \tag{1}$$

can be performed in a fairly straightforward way, with the help of the Gel'fand-Levitan integral equation, and practical algorithms have been developed.[3–7] In Eq. (1), $\Gamma(z)$ is the scattering-length density at a depth z from the surface determined by the product of the average nuclear scattering amplitude $b(z)$ and the atomic number density $N(z)$. If the phase is not known, least-squares-fit methods allow the determination of a depth profile,[8,9] but in general the solution is not unique.[10] In the literature, methods to retrieve the phase from the measurement of the reflectivity profile are discussed.[11] For one reflectivity profile these methods yield many solutions for the phase, stressing the nonuniqueness of the solutions. To decide which solution is correct, extra information is needed. This information could be knowledge of the structure and physics of the sample as deduced from either its preparation or its analysis by way of complementary depth-profiling techniques.[12] Another way to obtain more information about the sample is the use of contrast variation in neutron reflectively as obtained, for instance, by isotopic substitution.[13]

Phase information can be obtained by depositing the sample on a reference substrate, particularly if this is magnetized and the measurements are carried out with polarized neutrons.[14] While for an arbitrary orientation of the local magnetic field with respect to the neutron polarization axis the reflectivity has a slightly complex form, it simplifies when the two directions coincide.[15] When the magnetization is in the plane of the film, the neutrons experience a potential proportional to $(\Gamma\pm cB)$, where B is the magnetic induction and c is a material constant (here the z dependence is omitted for simplicity). The two signs within the parentheses refer to neutrons polarized parallel and opposite to the local magnetization, respectively. In writing this expression it has been assumed that the external magnetic field H may be neglected. Suppose that the sample is magnetized perpendicular to the plane of the film. The neutrons (regardless of their polarization) experience a potential proportional to Γ only, because in this geometry the solenoidal magnetic induction B is identical to the magnetic field H. Sivia and Pynn[16] and Majkrzak *et al.*[17] have already shown schemes for inverting neutron reflectivity data using polarized neutrons and a magnetized substrate. However, their method is based on the first Born approximation and it therefore holds only for values of q considerably larger than the critical value and small reflectivities.

Here a method is discussed to retrieve directly the phase information by measuring the reflectivity of an unknown layer through a known layer. From three measurements of the reflectivities of the unknown layer on three known layers having different reflectances, both the amplitude and the phase of the reflectance of the unknown layer can be calculated. The three known layers are obtained not by physical substitution, but by magnetizing the material in different configurations.

Assuming that the film on top of the substrate consists of two parts, whose reflecting properties are described by characteristic matrices F and G. This is shown schematically in Fig. 1. Part F is on top of G, so that the incident neutrons pass first through G and then F. The reflection and transmission amplitudes, r and t, respectively, are found from the matrix relation[18]

$$\begin{pmatrix} t \\ 0 \end{pmatrix} = M \begin{pmatrix} 1 \\ r \end{pmatrix} = FG \begin{pmatrix} 1 \\ r \end{pmatrix}, \tag{2}$$

and r takes the form

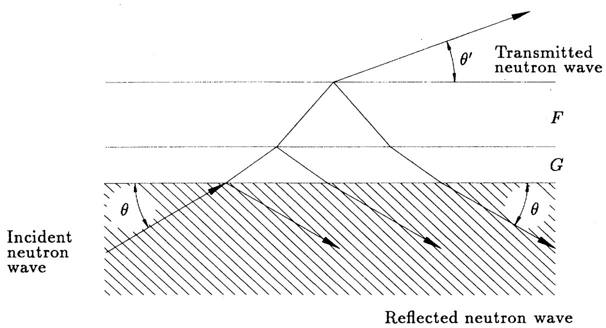

FIG. 1. Sketch of a neutron reflection experiment.

$$r=-\frac{m_{21}}{m_{22}}=-\frac{g_{11}r_f-g_{21}}{g_{12}r_f-g_{22}}\ , \tag{3}$$

where $r_f=f_{21}/f_{22}$. F and G describe the reflectances of the isolated films. The elements of G are known and those of F are unknown. When put together they might be thought of as separated by an infinitesimal (and irrelevant) gap. r_f is given by the expression

$$|r_f-r_c|^2=r_r^2\ , \tag{4}$$

where r_c and r_r are determined by the measured reflectivity $R=|r|^2$ and the known elements of the matrix G,

$$r_r=R^{1/2}\frac{|g_{11}g_{22}-g_{12}g_{21}|}{R\,|g_{12}|^2-|g_{11}|^2}\ ,$$
$$r_c=\frac{Rg_{22}g_{12}^*-g_{21}g_{11}^*}{R\,|g_{12}|^2-|g_{11}|^2}\ . \tag{5}$$

Equation (4) represents a circle in the complex reflectance plane with center at r_c and radius r_r. Both the amplitude and the phase of r_f can be uniquely determined from three measurements of R for three different values of G as the intersection of the three circles. Even if it is not formally proved, the physics involved demands that these circles have one common intercept. Two measurements (two circles) might suffice to retrieve the phase information; however, a third measurement gives the possibility to retrieve the phase information for all values of q independently. It helps also in removing any lingering ambiguity. The experimental value of R has finite precision, which means that the circumference of each circle has a certain thickness. The intersection of three circles pinpoints more precisely the real and imaginary parts of r_f.

In the following, results are presented of a numerical test. G, the magnetic layer, was taken to be a 10-nm-thick cobalt layer. This, however, had to be deposited on a material substrate and for this an infinitely thick layer of silicon was chosen. The "unknown" or test layer F was gold, 50-nm thick. All interfaces were taken to be sharp, except that at the surface (gold-air interface), which had a roughness of 1 nm. The neutrons enter the system from the silicon side. They encounter the sequence of scattering-length densities sketched in the inset of Fig. 2, with values derived from the bulk properties.

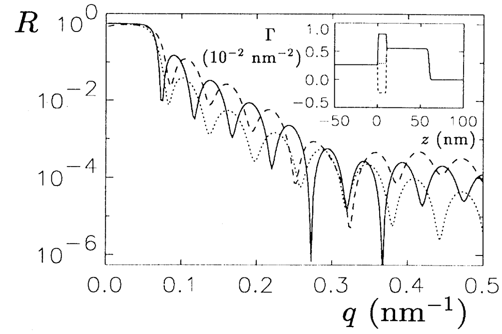

FIG. 2. Reflectivity of a sample consisting of a silicon substrate with a 10-nm-thick cobalt layer and a 50-nm-thick gold layer for three different magnetizations of the cobalt layer: not magnetized (dotted line) and plus (full line) and minus (dashed line) spins for a fully magnetized sample. The inset gives the scattering-length-density profiles for the three different measurements.

In Fig. 2 are presented the reflectivity for a nonmagnetized sample (dotted line) as well as the reflectivities for the fully magnetized sample with neutrons in the "plus" and "minus" spin states (respectively, continuous and dashed line). The retrieved amplitude and phase of the reflectance of F are shown as asterisks in Figs. 3 and 4, respectively. The continuous lines in the same figures are the results of the direct calculation of the reflectance starting from the neutron-optical potential of gold. Clearly the two results coincide. However, in the small-q region there are no values of the retrieved reflectance because the incident neutrons travel through silicon and the smallest q reachable is determined by the square root of the scattering-length density of silicon (0.051 nm^{-1}).

It was chosen to present the retrieved information as a reflectance rather than the scattering-length-density profile of the unknown layer. In the literature it has already been shown how this transformation can be performed in principle, and numerical results have been provided in a number of test cases.[7] The procedure requires values of the reflectance from $q=0$ to a maximum. In the case presented some sort of analytical continuation is needed to provide values of the reflectance below the minimal q experimentally reachable. An important aspect of the calculation is that it is performed for every q value independently. No correlation between the calculated reflectances is introduced. In practice this technique is complicated by the influence of statistics and resolution. It is possible that these effects, if large enough, interfere with the calculation of the mutual intersection of the circles. However, with sufficient statistics and resolution, this technique promises to be a powerful method for retrieving the scattering-length-density profile of nonmagnetic samples.

If the neutrons first pass the unknown film and then the known film, then the elements of G are unknown and those of F are known. The phase of $r_g=-g_{21}/g_{22}$ can not be determined because too many coefficients are unknown. In this case it is not possible to retrieve the exact

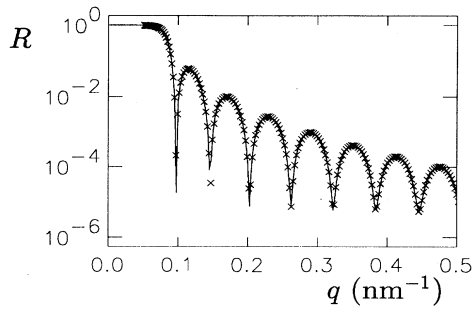

FIG. 3. Reflectivity of the gold layer, calculated from the simulated measurements of Fig. 2 (crosses) and using matrix calculations (line).

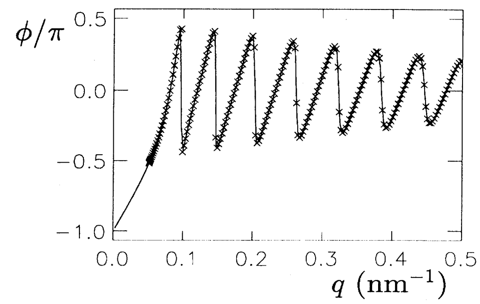

FIG. 4. Phase of the reflectance of the gold layer, ϕ, calculated from the simulated measurements of Fig. 2 (crosses) and using matrix calculations (line).

phase information for every q value independently without extra information about G with this method.

It was attempted by choice of test parameters to approach the conditions of a real experiment as closely as possible. Silicon is a substrate most readily available in a polished form. Cobalt can be easily deposited on it and is magnetically soft so that it can be saturated with reasonable fields both in the plane and out of the plane of the film. What remains to be done, of course, is a real experiment.

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