

# Complete determination of the reflection coefficient in neutron specular reflection by absorptive nonmagnetic media

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An experimental method is proposed which allows a complete determination of the complex reflection coefficient for absorptive media for positive and negative values of the momenta. It makes use of magnetic reference layers, and is a modification of a recently proposed technique for phase determination based on polarization measurements. The complex reflection coefficient resulting from a simulated application of the method is used for a reconstruction of the scattering density profiles of absorptive nonmagnetic media by inversion.

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## I. INTRODUCTION

Neutron specular reflection has become a common tool in materials sciences.<sup>1</sup> Nevertheless, there are still severe difficulties in the interpretation of reflectometer experiments because of the so-called *phase problem*. This problem refers to the incompleteness of the data obtained in standard experiments in structure physics,<sup>2,3</sup> where only the intensities of the reflected waves are measured but not the corresponding phases. In specular reflection both the modulus and the phase of the reflection coefficient are needed for an unambiguous reconstruction of the surface profiles.<sup>4,5</sup>

Several solutions of this so-called phase problem have been proposed for neutron specular reflection.<sup>4-13</sup> Recently proposed reference layer methods based on polarization measurements<sup>9,10</sup> are of particular interest, because they also work in the total reflection regime, and allow unique reconstruction of surface profiles of magnetic samples. The experimental implementation of the reference layer method of Majkrzak and Berk<sup>7</sup> was first achieved with good success by Majkrzak *et al.*,<sup>14</sup> who also proposed and tested experimentally the related *surround method*.<sup>15,16</sup>

The available methods of phase determination (e.g., Refs. 7-16), for nonabsorptive as well as absorptive samples, provide us with the full (modulus and phase) reflection coefficient  $R(q)$  for positive values of the wave number  $q$  perpendicular to the surface of the sample. On the other hand, the reconstruction of the surface profiles by inversion requires a knowledge of  $R(q)$  over the positive *and* negative range of  $q$  values. This is no problem for nonabsorptive samples, because in this case the reflection coefficient  $R(q)$  satisfies

$$R(-q) = R^\dagger(q), \quad (1)$$

and  $R(q)$  at negative  $q$  values is directly given in terms of  $R(q)$  at positive  $q$ . However, in the presence of absorption, relation (1) does not hold, and therefore a reconstruction of the surface profiles is not possible without further input. To our knowledge, so far no procedure has been proposed for providing the missing information.

In this work we propose a modification of the reference layer method for nonmagnetic samples based on polarization measurements.<sup>9</sup> The modification consists of an interchange of the positions of the sample and the reference layer, which allows a determination of the reflection coefficient for positive and negative  $q$  values. The method also works in the total reflection regime (in contrast to Refs. 7,8); it is only limited by the sample thickness because of the corresponding Kiessig oscillations which must be resolved.

In Sec. II we present the method, and derive the basic relations for the phase determination. A realistic example is given in Sec. III, where by simulations we demonstrate the feasibility of the method. The problem of thick samples is also discussed. A brief summary and concluding remarks are given in Sec. IV.

## II. THE METHOD

We consider the arrangement of an unknown nonmagnetic sample and a magnetic reference layer mounted on a substrate (e.g., a Si wafer), as shown in Fig. 1. We assume that within the reference layer ( $0 \leq x \leq a$ ) there is a magnetic field  $\mathbf{B}(x)$  aligned with the surface of the sample. The direction of this field is taken as the  $z$  axis, which is chosen as the axis of spin quantization. The direction of propagation perpendicular to the surface defines the  $x$  axis in a right-handed coordinate system. Such a magnetic reference layer may consist of a ferromagnetic stratum, e.g., a Fe, Co, or Ni layer (cf. Fig. 1). We assume that the sample as well as the substrate are field free. However, further considerations still remain valid as long as the condition  $|\mathbf{B}(x)| = 0$  for  $x \rightarrow \infty$  is satisfied.

The arrangement of Fig. 1 differs from that of Ref. 9 only by the interchange of the sample and reference layer, and therefore we can make use of the same relationships for a description of the reflection. The position of the sample on top of the reference layer and the substrate, however, leads to a completely new situation as compared to previous proposals for the solution of the phase problem. This is easily seen

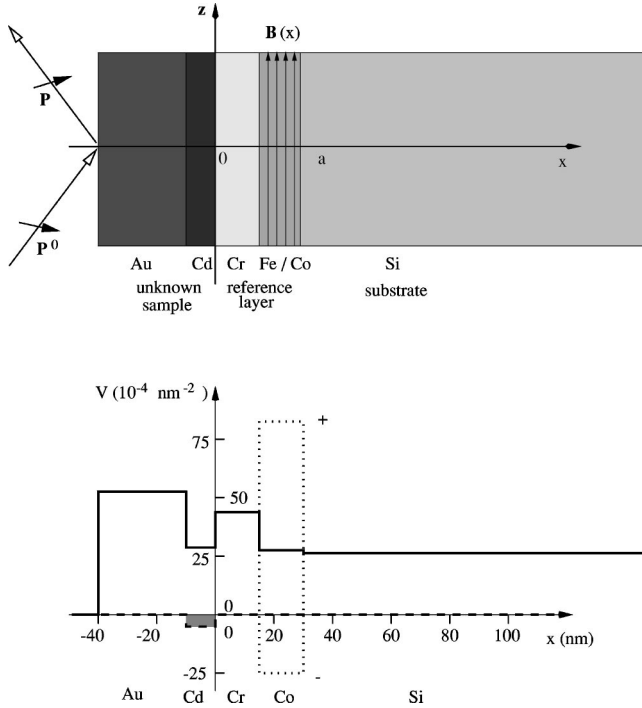


FIG. 1. Experimental arrangement for measuring the complex reflection coefficient. Top: arrangement of the layers. Bottom: the potential profile; the real part is represented by the solid lines, and the imaginary part by the dashed area. The dotted lines represent the effective potentials experienced by neutron beams polarized parallel and antiparallel to the magnetic field  $\mathbf{B}$ .

from the expression for the reflection coefficient  $R^{tot}$  of the whole arrangement:

$$R_L^{tot}(q) = \frac{E(q)\rho_L(q) + R_L(q)}{1 - R_R(q)\rho_L(q)}. \quad (2)$$

Here the reflection properties of the sample enter in terms of the reflection coefficients  $R_L$  and  $R_R$  and the quantity

$$E(q) = T_L(q)T_R(q) - R_L(q)R_R(q), \quad (3)$$

where  $T_L$  and  $T_R$  are the transmission coefficients of the sample. The quantity  $\rho_L$  is the reflection coefficient of the reference layer plus the substrate, and the indices  $L$  and  $R$  refer to incidence from left and right, respectively. Because of the rather intricate dependence of  $R_L^{tot}$  on the reflection properties of the sample, the separation of its unknown reflection coefficient becomes more involved.

Following the procedure of Ref. 9, we introduce the quantity

$$s = \frac{R_{L+}^{tot}}{R_{L-}^{tot}}, \quad (4)$$

which can be expressed in the form

$$s = \frac{P_x^0 + iP_y^0}{P_x + iP_y} \frac{1 + P_z}{1 + P_z^0}, \quad (5)$$

where  $\mathbf{P} = (P_x, P_y, P_z)$  and  $\mathbf{P}^0 = (P_x^0, P_y^0, P_z^0)$  are the polarizations of the reflected and the incident beam, respectively. The indices  $\pm$  refer to neutron beams polarized parallel (+) and antiparallel (-) to the magnetic field. All quantities on the right-hand side of Eq. (5) are measurable, and therefore  $s$  can be determined experimentally.

Use of Eq. (2) for  $R_{\pm}^{tot}$  in Eq. (4) leads, after simple algebraics, to the relation

$$-\frac{R_L(q)}{E(q)} = \frac{(\rho_L^+ - s\rho_L^-) - R_R\rho_L^+\rho_L^-(1-s)}{(1-s) - R_R(\rho_L^- - s\rho_L^+)}. \quad (6)$$

The term on the left-hand side of Eq. (6) is equal to  $R_R(-q)$ , i.e., to the right reflection coefficient at negative  $q$  values (cf. Refs. 17,18), and therefore Eq. (6) becomes

$$R_R(-q) = \frac{(\rho_L^+ - s\rho_L^-) - R_R(q)\rho_L^+\rho_L^-(1-s)}{(1-s) - R_R(q)(\rho_L^- - s\rho_L^+)}. \quad (7)$$

This is a relation involving measurable quantities which expresses the right reflection coefficient at  $-q$  ( $q > 0$ ) in terms of its value at  $q$ .

For a full determination of the reflection coefficient  $R_R(q)$  at positive and negative  $q$  values [i.e., for obtaining two independent equations for the two unknowns  $R_R(q)$  and  $R_R(-q)$ ], at least two sets of measurements with different reference layers are necessary. Denoting the two measurements with the upper indices ( $a$ ) and ( $b$ ), from Eq. (7) one obtains the quadratic equation for  $R_R(q)$ ,

$$\alpha + \beta R_R(q) + \gamma R_R^2(q) = 0, \quad (8)$$

with

$$\alpha = (1 - s^{(b)})(\rho_L^{+(a)} - s^{(a)}\rho_L^{-(a)}) - (1 - s^{(a)})(\rho_L^{+(b)} - s^{(b)}\rho_L^{-(b)}), \quad (9)$$

$$\begin{aligned} \beta = & (\rho_L^{-(b)} - s^{(b)}\rho_L^{+(b)})(\rho_L^{+(a)} - s^{(a)}\rho_L^{-(a)}) \\ & - (\rho_L^{+(b)} - s^{(b)}\rho_L^{-(b)})(\rho_L^{-(a)} - s^{(a)}\rho_L^{+(a)}) \\ & + (1 - s^{(a)})(1 - s^{(b)})\rho_L^{+(a)}\rho_L^{-(a)} \\ & - (1 - s^{(a)})(1 - s^{(b)})\rho_L^{+(b)}\rho_L^{-(b)}, \end{aligned} \quad (10)$$

$$\begin{aligned} \gamma = & (1 - s^{(a)})(\rho_L^{-(b)} - s^{(b)}\rho_L^{+(b)})\rho_L^{+(a)}\rho_L^{-(a)} - (1 - s^{(b)}) \\ & \times (\rho_L^{-(a)} - s^{(a)}\rho_L^{+(a)})\rho_L^{+(b)}\rho_L^{-(b)}. \end{aligned} \quad (11)$$

The quadratic equation (8) has two roots

$$R_R^{(1,2)}(q) = \frac{-\beta \pm \sqrt{\beta^2 - 4\alpha\gamma}}{2\gamma}. \quad (12)$$

Similarly to the procedure of Ref. 9, the physical solution  $R_R(q)$  can be selected either by continuity requiring that the phase  $\phi(q)$  satisfies  $\phi(q=0) = -\pi$  or by the condition  $r_R = |R_R|^2 \leq 1$ . The latter may fail at some  $q$  values, and must be accompanied by continuity in certain momentum regions. The selection by continuity implies measurements of  $s$  over

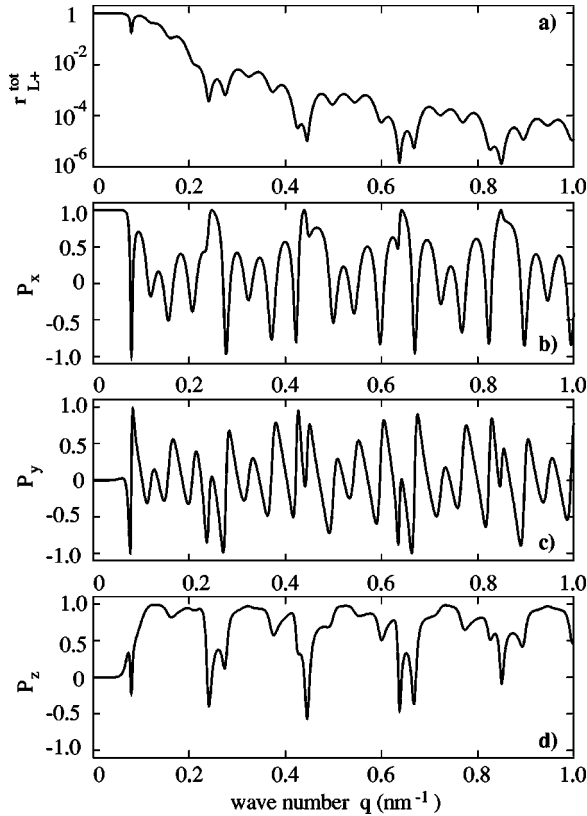


FIG. 2. Simulated reflectivity and polarization data for the arrangement of Fig. 1 with a magnetized Fe film in the reference layer. The incident beam is assumed to be fully polarized in the  $x$  direction. The reflectivity  $r_{L+}^{tot} = |R_{L+}^{tot}|^2$  (a) and the polarization components  $P_x$  (b),  $P_y$  (c), and  $P_z$  (d) of the reflected beam are shown.

the whole range of momenta. The solution  $R_R(-q)$  at negative  $q$  values is given by Eq. (7).

We note that the method relies solely on measurements of the polarization of the reflected beam. In particular, even for absorptive samples, no transmission measurements are needed for the determination of  $R(-q)$ , as might be conjectured from the general formalism.<sup>18</sup>

### III. EXAMPLE

We test the method by a simulation using the realistic example shown in Fig. 1. The sample consists of a 30-nm Au layer and a 10-nm Cd layer, where the latter is strongly absorptive. To determine the reflection coefficient for positive and negative  $q$  values, we consider measurements with two different reference layers. Both reference layers are composed of a 15-nm Cr layer and a 15-nm-thick film of ferromagnetic material magnetized up to saturation and mounted on a Si wafer. The required difference in the reflection properties of the reference layers can be achieved by using in the measurements Fe (a) and Co (b) for the ferromagnetic layer. The magnetization of this layer will also generate a magnetic induction outside the ferromagnetic film, which we assume,

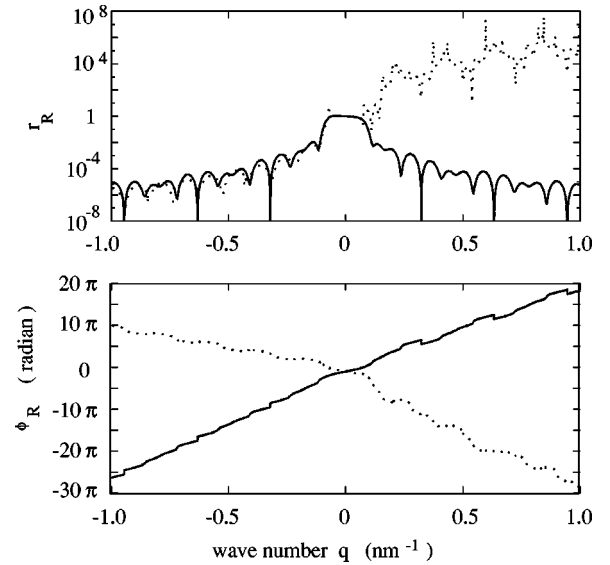


FIG. 3. The roots  $R_R^{(1,2)}$  of the quadratic equation as obtained from the measurements of the polarization components. Top: the reflectivities  $r_R^{(1,2)} = |R_R^{(1,2)}|^2$ . Bottom: the absolute reflection phases  $\phi_R^{(1,2)}$  are shown assuming  $\phi_R = -\pi$  at  $q=0$ . The solid curves correspond to the physical solution.

however, to be small enough not to affect the neutron beam. For simplicity, we set it equal to zero.

The reflectivity  $r_{L+}^{tot} = |R_{L+}^{tot}|^2$  and the polarization components  $P_x, P_y$ , and  $P_z$  for the arrangement of Fig. 1 have been calculated for both reference layers at all positive  $q$  values. To illustrate the typical observables which have to be measured for phase determination, we show in Fig. 2 the reflectivity  $r_{L+}^{tot} = |R_{L+}^{tot}|^2$  and the components of the polarization  $P_x, P_y$ , and  $P_z$  for the reference layer (a), assuming a fully polarized incident beam in the  $x$  direction. It is evident that in the given example measurements in the range  $0 \leq q \leq 1.0 \text{ nm}^{-1}$  are certainly feasible.

Following the procedure outlined in Sec. II, we obtain two solutions for  $R_R(q)$  which are displayed in Fig. 3. As expected, in a wide range of positive  $q$  values only one solution satisfies  $|R_R(q)| \leq 1$  and is therefore physically admissible. Applying continuity, the region of uniqueness can be extended over all positive  $q$  values. In addition, via Eq. (7) one obtains unique values of  $R_R(-q)$ . Hence the measurements of the polarization at positive  $q$  values are sufficient for a determination of the complete reflection coefficient on the whole  $q$  axis. To extract the surface profiles from the extracted reflection coefficients  $R_R(q)$ , we apply the Marchenko inversion procedure used in Ref. 17, where it was shown to yield correct reconstructions of the *complex* input potential for some examples modeled on realistic cases (iron and silver layers on glass), although a rigorous mathematical proof of the validity of this procedure is lacking. We regard this as acceptable from a physicist's point of view. The Marchenko inversion procedure is usually formulated for  $R_L(q)$ , while our procedure provides  $R_R(q)$ . Because of the specific choice of our coordinate system we can also use  $R_R(q)$  in the formulation of Ref. 17 but we have to change

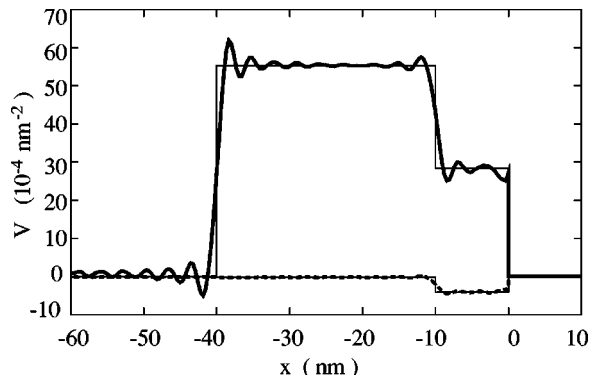


FIG. 4. The potential profile of the sample obtained by inversion of the extracted values of the reflection coefficient in the momentum range  $-1.0 \leq q \leq 1.0 \text{ nm}^{-1}$  shown in Fig. 3. The real and imaginary parts of the reconstructed profile are shown by thick solid and dashed lines, respectively. For reference, the original profile is shown by thin solid lines.

the argument of the evaluated potential from  $x$  to  $-x$ . This procedure has been applied to analyze the numerically extracted values of  $R_R(q)$ . The results for the scattering length density profile are displayed in Fig. 4. It is obvious that the method reproduces the original potential within the resolution determined by the maximum available  $q$  value.

Since the method is strongly related to the reference layer method presented previously,<sup>9</sup> its stability with respect to experimental uncertainties, e.g., roughness of interfaces and measurement errors, is similar to that of the method of Ref. 9. This is also true for the reconstruction of the surface profiles. For more details on the stability we refer to our previous studies (cf. Refs. 9 and 19).

#### IV. CONCLUSIONS

We have proposed a method involving a magnetic reference layer which allows a determination of the full complex reflection coefficient at positive and negative  $q$  values. Thus a reconstruction of surface profiles of absorptive nonmagnetic samples becomes feasible. The procedure is a modification of the previously proposed method.<sup>9</sup> The main difference is the interchange of the position of the sample and the reference layer. Because of this change the reflection properties of the sample enter in a more involved way, and require two sets of measurements using two different reference layers. The additional effort is remunerated by providing us with an additional relationship from which the reflection coefficient at negative  $q$  values can be obtained. This feature allows a determination of absorptive surface profiles by inversion—an open problem up to now. In a numerical example, it is demonstrated that the reconstruction of real and imaginary parts of the scattering density profiles is feasible.

As in the previously proposed reference layer methods,<sup>7-9</sup> the thickness of the layers is reflected in the  $q$  dependence of the measured data. Specifically, the so-called Kiessig oscillations must be resolved in experiment. Hence the degree of the monochromaticity, as well as the resolution of the diffraction angle of the incident beam, set a limit on the thickness of the studied system. The roughness of the interfaces has similar effects and results in additional uncertainties in the deduced reflection coefficient (cf. Ref. 9).

In summary the phase problem of neutron reflection has been solved for absorptive non-magnetic samples. The method seems experimentally feasible for sample thicknesses up to 100 nm, and complements reference layer methods<sup>9,10</sup> recently presented.

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