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Unique reconstruction of depth profiles in neutron specular reflectometry: practical aspects

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Abstract

The possibility of a full determination of the reflection coefficient (modulus and phase) in neutron specular reflection is discussed. Specifically, the proposed solution of the phase problem based on polarization measurements and a magnetic reference layer is considered. A procedure and the corresponding formulae are worked out to extract the maximum of accessible information from measurements with available reflectometers, taking into account their limitations in the polarization analysis and the preparation of the beam. Albeit the incompleteness of the polarization measurements results in ambiguities of the reflection coefficient, the physical one can in most cases be identified by additional knowledge about the profile. This is shown in a schematic example with simulated incomplete data.
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1. Introduction

Spin-polarized neutron specular reflection has become an important tool for the study of thin films and superlattices [1]. It not only provides essential information on the nuclear scattering length density profile, but also on the magnetization profile of a stratified sample. The unique interpretation of the reflection data in terms of the scattering length profile requires the full knowledge of the reflection coefficient (modulus and phase). In general, the phases of the reflection matrix are not obtained in standard reflectometry. Several proposals to solve this so-called phase problem have been reported. At present, the methods which make use of the spin-dependent interaction of a neutron with a magnetic reference layer [2–5] are most promising. Although they require only minor modifications of the standard setup, solely the method of Majkrzak and Berk [5] has been tested experimentally [6] for a specific case.

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As outlined in Ref. [3] a complete set of polarization measurements is an important step in the solution of the phase problem because it allows the determination of the reflection matrix $\mathcal{R}$ up to one unknown phase (e.g. φ_{++})

$$\mathcal{R}(q) = \begin{pmatrix} R_{++} & R_{+-} \\ R_{-+} & R_{--} \end{pmatrix} = |R_{++}| e^{i\varphi_{++}} \begin{pmatrix} 1 & g_{+-} \\ g_{-+} & g_{--} \end{pmatrix}, \qquad (1)$$

where the indices refer to the up (+) and down (−) spin components and q is the wave number perpendicular to the surface of the sample. The matrix elements $g_{+-} = R_{+-}/R_{++}$, $g_{-+} = R_{-+}/R_{++}$ and $g_{--} = R_{--}/R_{++}$ are complex and can be fully determined via a three-dimensional polarization analysis of the reflected beam [3,7]. However, at present the available reflectometers are limited with regard to the choice of the polarization of the incident beam as well as in the polarization analysis of the reflected beam. Therefore, one can perform only an incomplete polarization analysis of the reflected beam and one wonders about the consequences for the retrieval of the profiles.

In this contribution, we aim at a method to extract the maximum of information on the matrix elements g_{ij} with such a restricted polarization analysis. In Section 2, we outline the method and derive relationships up to which the matrix elements g_{ij} can be determined in such a set-up. In Section 3, we consider a schematic example of a non-magnetic sample and find that the ambiguities in the analysis due to incomplete polarization measurement does not hamper the unique retrieval of the profile.

2. Incomplete polarization measurements

In the original proposal of Kasper et al. [3], it is assumed that a full three-dimensional polarization analysis of the reflected beam can be performed. In addition, there should be also the possibility to rotate the polarization of the incident beam. Albeit technically feasible, the presently available reflectometer set-ups are limited with regard to such a handling of the polarization. In general, the incident beam can be polarized only in one direction and the polarization analysis of the reflected beam is usually limited to the component in this direction. Hence, the set-ups are designed for measurements of the reflectivities $r_{\pm\pm} = |R_{\pm\pm}|^2$ and $r_{\pm\mp} = |R_{\pm\mp}|^2$, but not for the determination of the polarization. Due to these restrictions, a specific set of measurements and corresponding analysis is required to extract also some phase information on the matrix elements g_{+-}, g_{-+} and g_{--}.

Following the procedures of quantum mechanics the density matrix of the reflected beam is given by $\rho = \mathcal{R}\rho_0\mathcal{R}^\dagger/\mathrm{Trace}(\mathcal{R}\rho_0\mathcal{R}^\dagger)$, where $\rho_0 = \frac{1}{2}(1 + \boldsymbol{\sigma} \cdot \mathbf{P}_0)$ is the density matrix of the incident beam which is given in terms of its polarization $\mathbf{P}_0$. The polarization of the reflected beam is then given by $\mathbf{P} = \mathrm{Trace}(\boldsymbol{\sigma} \cdot \rho)$.

For a first group of measurements, we align the y-axis with the polarization of the incident beam ($P_x^0 = P_z^0 = 0$) and use the z-axis for quantization as usual. Straightforward algebra yields

$$P_y = \frac{a + bP_y^0}{\Gamma + cP_y^0} \quad \text{with}$$

$$\Gamma = \frac{r_{++} + r_{+-} + r_{-+} + r_{--}}{r_{++}} \qquad (2)$$

and the real quantities

$$\begin{aligned} a &= -\mathrm{i}[g_{-+} - g_{-+}^* + g_{--}g_{+-}^* - g_{+-}g_{--}^*], \\ b &= [g_{--} + g_{--}^* - g_{-+}g_{+-}^* - g_{+-}g_{-+}^*], \\ c &= +\mathrm{i}[g_{+-} - g_{+-}^* - g_{--}g_{-+}^* + g_{-+}g_{--}^*], \end{aligned} \qquad (3)$$

where $*$ denotes complex conjugation. It is obvious that the measurement of the y-component of $\mathbf{P}$ contains part of the information on the phases of the reflection matrix $\mathcal{R}$. Performing three measurements with different polarization P_y^0, e.g. $P_y^0 = -1, 0$ and $+1$, allow the determination of the three quantities a, b and c.

For the determination of Γ, we need a second group of measurements in which the direction of the polarization of the incident beam is rotated. Since most standard reflectometers do not allow for such a rotation of the polarization of the incident beam, one can achieve the same effect, if one rotates the sample by 90° with the rotation axis aligned with the x-direction. Thus the axes are $\hat{\mathbf{y}}' = -\hat{\mathbf{z}}$, $\hat{\mathbf{z}}' = \hat{\mathbf{y}}$ and $\hat{\mathbf{x}}' = \hat{\mathbf{x}}$. Effectively, one measures then P_z of the reflected beam for an incident beam directed in $-z$-direction. Using the same formalism as in Eq. (2), one obtains

$$P_z = -\frac{[r_{++} + r_{+-} - r_{-+} - r_{--}] - [r_{++} - r_{+-} - r_{-+} + r_{--}]P_z^0}{[r_{++} + r_{+-} + r_{-+} + r_{--}] - [r_{++} - r_{+-} + r_{-+} - r_{--}]P_z^0}. \quad (4)$$

Performing again three measurements, e.g. $P_z^0 = -1, 0, +1$ allows the extraction of $|g_{+-}|$, $|g_{-+}|$ and $|g_{--}|$ and therefore the determination of Γ. As can be seen from Eq. (4), the polarization P_z does not contain phase information. Therefore, the determination of the polarization is not required and it suffices to measure the reflectivities $r_{\pm\pm}$ and $r_{\pm\mp}$.

After the determination of the quantities a, b and c from the first set of measurements and assuming that we know the reflectivities $r_{\pm\pm}$ and $r_{\pm\mp}$ we can write down a set of equations for the phases γ_{ij} ($g_{ij} = |g_{ij}| \exp(\mathrm{i}\gamma_{ij})$),

$$\begin{aligned} a &= 2|g_{-+}| \sin\gamma_{-+} + 2|g_{-+}||g_{--}| \sin(\gamma_{--} - \gamma_{+-}), \\ b &= 2|g_{--}| \cos\gamma_{--} - 2|g_{-+}||g_{+-}| \cos(\gamma_{-+} - \gamma_{+-}), \\ c &= -2|g_{+-}| \sin\gamma_{+-} - 2|g_{-+}||g_{--}| \sin(\gamma_{--} - \gamma_{-+}). \end{aligned} \quad (5)$$

We did not find a closed solution of this set of coupled non-linear equations for phases γ_{ij}. Therefore, one has to apply numerical means for the determination of the phases. This has the drawback that we cannot give a general statement on the ambiguities of the analysis due to the incomplete polarization measurement. The phases γ_{ij} enter Eqs. (5) only as arguments of trigonometric functions. Therefore, we conjecture from the specific case below that there will be only a small number of discrete ambiguities (probably only two solutions).

The situation becomes particularly simple for a sample which is magnetized in z-direction only, i.e. $\mathbf{B}(x) = B(x)\hat{\mathbf{z}}$. In this special case the off-diagonal matrix elements of $\mathscr{R}$ vanish, i.e. $R_{\pm\mp} = 0$. Hence, Eqs. (2) and (4) reduce to

$$P_y = \frac{g_{--}^* + g_{--}}{1 + g_{--}g_{--}^*} P_y^0, \quad (6)$$

$$P_z = \frac{[r_{++} - r_{--}] + [r_{++} + r_{--}]P_z^0}{[r_{++} + r_{--}] + [r_{++} - r_{--}]P_z^0}. \quad (7)$$

Performing a first reflection experiment with an unpolarized incident beam allows the determination of $|g_{--}|^2$ from a measurement of the polarization component aligned with the magnetization of the sample. This standard experiment has to be supplemented by a second experiment in which the sample is rotated by 90°, so that the incident polarization as well as the measured polarization component is perpendicular to the magnetization direction of the sample ($P_y^0 \neq 0, P_x^0 = P_z^0$, sample magnetized in z-direction). From Eq. (6) we can extract an equation for the phase of g_{--}

$$\cos\gamma_{--} = \frac{1 + |g_{--}|^2}{2|g_{--}|} \frac{P_y}{P_y^0}. \quad (8)$$

Because of the properties of the cosine function, the phase γ_{--} cannot be extracted uniquely from Eq. (8), thus leading to an ambiguity in the sign of $\sin\gamma_{--}$.

It should be noted that the same expressions for P_y and P_z are obtained via the co-ordinate independent formalism of Rühm et al. [8] if the representation of $\mathscr{R}$ given in Eq. (1) is used.

3. Example

The incomplete polarization analysis in the reflected beam leads to an ambiguity in the analysis of the phase and, therefore, also in the reconstructed depth profile of the sample. In order to investigate this problem for the profile retrieval, we consider a schematic example. For simplicity we restrict ourselves to the case of a non-magnetic sample and a simple reference layer magnetized only in one direction. The sample consists of a 15 nm Au layer. On top of the sample, a 20 nm Fe-layer magnetized up to saturation is used as a reference layer.

We simulate the reflection data for the total arrangement, i.e. sample plus reference layer, where the incident beam is polarized in y-direction and the magnetization of the reference layer is in z-direction. We assume that in a previous measurement $|g_{--}|$ was already determined (see Section 2). Via Eq. (8) we can determine $\cos\gamma_{--}$ and, therefore, $\sin\gamma_{--}$ up to the sign. Hence, we cannot distinguish between g_{--} and g^*_{--} due to the incomplete polarization measurement.

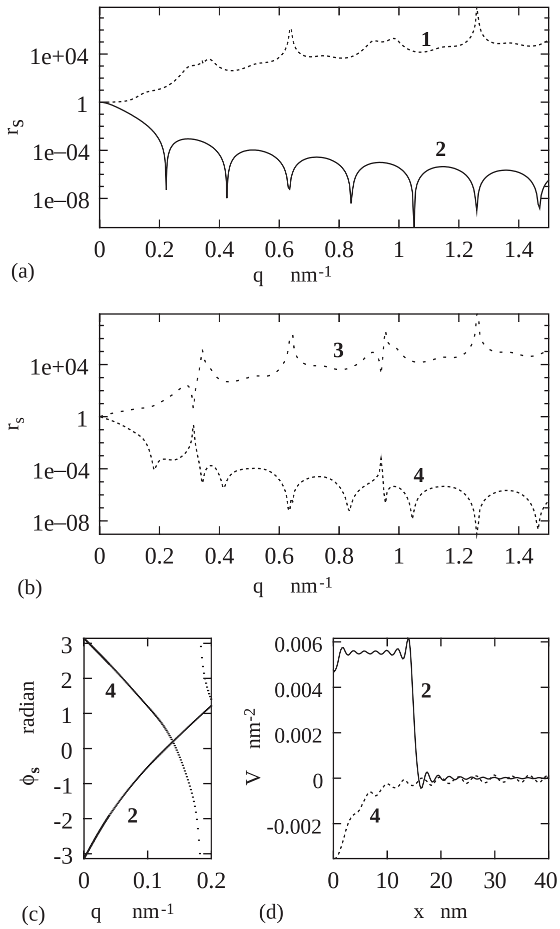

Fig. 1. Reconstruction of the example profile from simulated reflection data. Details are explained in Section 3: (a) first and second solution of the reflectivity r_s; (b) third and fourth solution of the reflectivity r_s; (c) the second and third solution of the phase ϕ_s in the vicinity of $q = 0$; (d) reconstructed scattering length density profile of the second and the fourth solution.

Following the procedure outlined in Ref. [2], we have used the two values g_{--} and g^*_{--} as two possible values of $s = R_{++}/R_{--} = 1/g_{--}$ Ref. [2] Eq. (10) and determined the reflection coefficient $R_s = \sqrt{r_s}\exp(i\phi_s)$ of the sample. In principle one obtains four solutions for R_s shown in Fig. 1. As already outlined in Ref. [2], two solutions are unphysical because they lead to reflectivities $r_s > 1$. From the remaining two solutions only one is physical because it has the correct behaviour $\phi_s \to -\pi$ for $q \to 0$. This is also seen in a reconstruction of the surface profile of the sample from R_s via inverse scattering methods [9]. From the extracted physical R_s one obtains within the accuracy of integration, the original Au-layer in depth and width. Using the unphysical solution leads to a completely unrealistic profile.

In summary, we have shown that the incomplete polarization analysis in standard reflectometers lead to an ambiguity in the extracted reflection matrix of the sample. There are good reasons that the number of ambiguous solutions for the reflection matrix is small and one may be able to select the physical one because most solutions will exhibit unphysical properties. Our considerations clearly indicate that measurements at available reflectometers with limited polarization analyses may allow unique retrievals of depth profiles, although the corresponding phase determination is ambiguous.

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