Phase Determination in Spin-Polarized Neutron Specular Reflection

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(Received 6 November 1997)

New spin-polarized neutron specular reflection experiments to determine the moduli and phases of the full reflection matrix (non-spin-flip and spin-flip amplitudes) are proposed. The method makes use of a reference layer and exploits the interference of the spin components of the neutron beam as a function of incident polarization. [S0031-9007(98)05598-7]

PACS numbers: 61.12.Ha, 28.20.–v

Spin-polarized neutron specular reflection is increasingly used for the study of spin structures in thin magnetic films and superlattices [1]. It provides important information on the magnetic properties in nanostructured materials, such as magnetization profiles and magnetic coherence lengths. Several instruments for neutron specular reflection studies are available worldwide which provide the option for polarized beam scattering and analysis.

In specular reflection one is dealing with one-dimensional quantal scattering by a potential $\mathcal{V}$ which depends only on the coordinate perpendicular to the surface. $\mathcal{V}$ is composed of a nuclear part, $\mathcal{V}_{\rm nucl}$, proportional to the scattering-length density profile, and a magnetic part, $\mathcal{V}_{\rm magn} = \gamma\boldsymbol{\sigma}\cdot\mathbf{B}$, where γ is the gyromagnetic factor of the neutron, $\mathbf{B}$ is the magnetic field, and $\boldsymbol{\sigma}$ is the vector of the standard Pauli matrices.

The magnetic term couples the spin components and, therefore, one has to consider a coupled one-dimensional Schrödinger equation with the interaction

$$\mathcal{V} = \begin{pmatrix} V_{\rm nucl} + \gamma B_z & \gamma B_x - i\gamma B_y \\ \gamma B_x + i\gamma B_y & V_{\rm nucl} - \gamma B_z \end{pmatrix}. \qquad (1)$$

The nuclear part, $V_{\rm nucl}$, is, in general, the same for both spin components. The reflection of a polarized neutron beam with normal wave number q is described by the 2×2 reflection matrix $\mathcal{R}(q)$. The diagonal elements correspond to the non-spin-flip coefficients; the nondiagonal elements describe spin-flip processes. Similarly, one has a 2×2 transmission matrix $\mathcal{T}(q)$.

In actual spin-polarized neutron specular reflection experiments [1], only the moduli of the matrix elements of $\mathcal{R}$ are usually measured via the reflectivities. These reflectivity data can be analyzed only in a model-dependent way involving fitting procedures because an unambiguous retrieval of the scattering-length and magnetic field profiles requires the knowledge of the full reflection matrix, namely, the moduli and phases of all matrix elements. This is the so-called *phase problem* which has been widely discussed in standard ("spinless") reflectometry with neutrons and x rays, where it has hampered the application of the method considerably.

Recently, several schemes for the determination of the phase in standard neutron specular reflection experiments have been proposed. We mention three approaches: (i) the reference layer method where one makes use of the interference between the reflections of a known reference layer and the unknown surface profile [2–6], (ii) the Lloyd's mirage technique [3], and (iii) the dwell time method [7]. Among these the reference layer methods are most promising because they require only minor modifications of the usual setup. Although so far only one proposal has been implemented in experiment [8] it is fair to say that the phase problem of standard neutron specular reflection is now solved in principle. In spin-polarized neutron specular reflection, however, the determination of the phases is still an open problem and we are not aware of any attempt of its solution.

In this Letter we propose a novel method for the measurement of the full reflection matrix in spin-polarized neutron specular reflection. Measurements of the polarization of the reflected beam and use of a reference layer are required. For a full determination of $\mathcal{R}$, measurements with differently polarized incident beams are needed. As shown by tests with simulated data the procedure is stable against experimental uncertainties.

We consider a neutron reflectometer setup which allows for variations of the polarization directions of the incident beam as well as polarization measurements of the reflected beam. This setup is the same as that in our recent proposal for the phase determination in standard neutron reflectometry [6]. The procedure for the phase determination, however, is different because of the presence of magnetic fields in the sample.

We describe the reflection process in the general formalism of the density matrix ρ which for a neutron beam is a 2×2 matrix in spin space. The Pauli matrices $\boldsymbol{\sigma}$ together with the unit matrix form a basis in this space so we can write the density matrix ρ^0 of the incident neutron beam in the form [9]

$$\rho^0 = \tfrac{1}{2}(1 + \mathbf{P}^0\cdot\boldsymbol{\sigma}) \qquad (2)$$

with the polarization vector

$$\mathbf{P}^0 = \mathrm{Tr}(\rho^0\boldsymbol{\sigma}). \qquad (3)$$

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The density matrix of the reflected beam is then

$$\rho = \mathcal{R}\rho^0\mathcal{R}^+, \tag{4}$$

where the reflection matrix

$$\mathcal{R} = \begin{pmatrix} R_{++} & R_{+-} \\ R_{-+} & R_{--} \end{pmatrix} \tag{5}$$

is defined in an arbitrarily chosen system of spin quantization. The polarization vector of the reflected beam is given by

$$\mathbf{P} = \mathrm{Tr}(\rho\boldsymbol{\sigma})/\mathrm{Tr}(\rho). \tag{6}$$

We now define the quantity

$$s_{\hat{g}} = \left.\frac{P_+}{1 + P_z}\right|_{\hat{g}}, \tag{7}$$

where P_+ denotes the combination $P_+ = P_x + iP_y$ and the lower index $\hat{g}$ refers to an incident beam which is fully polarized parallel to $\hat{g}$. Using (4) to (6) for differently polarized incident beams we find

$$\begin{aligned} s_{\pm\hat{x}} &= \frac{R_{-+} \pm R_{--}}{R_{++} \pm R_{+-}}, & s_{\pm\hat{y}} &= \frac{R_{-+} \pm iR_{--}}{R_{++} \pm iR_{+-}}, \\ s_{+\hat{z}} &= \frac{R_{-+}}{R_{++}}, & s_{-\hat{z}} &= \frac{R_{--}}{R_{+-}}. \end{aligned} \tag{8}$$

As can be seen from Eqs. (7) and (8) the measurement of $\mathbf{P}$ determines ratios of the matrix elements of $\mathcal{R}$. There are three independent ratios (e.g., $R_{-+}/R_{++}, R_{+-}/R_{++}, R_{--}/R_{++}$), and we must choose three of the six relations (8) (corresponding to three choices of the polarization of the incident beams) to calculate them. This leads to a system of three linear equations for the three unknown ratios. The different ways of determining these ratios are mathematically equivalent but result in a different behavior in regard to the propagation of experimental errors.

The reflection matrix $\mathcal{R}$ can now be written in the form

$$\mathcal{R} = R_{++}\mathcal{G}, \tag{9}$$

where the matrix

$$\mathcal{G} = \begin{pmatrix} 1 & R_{+-}/R_{++} \\ R_{-+}/R_{++} & R_{--}/R_{++} \end{pmatrix} \tag{10}$$

is completely determined by the measurement of the polarization of the reflected beams for three differently polarized incident beams. We are left with the problem of the determination of *one* matrix element of $\mathcal{R}$. We have chosen R_{++}, but equivalently one may take any other matrix element.

To determine R_{++} we propose a method which involves two types of measurements. First, one performs the polarization measurements, as discussed above, with the sample alone. These yield $\mathcal{G}^{\mathrm{S}}$, where the upper index "S" refers to the sample. Second, one places a known reference layer on top of the sample (see Fig. 1) and repeats

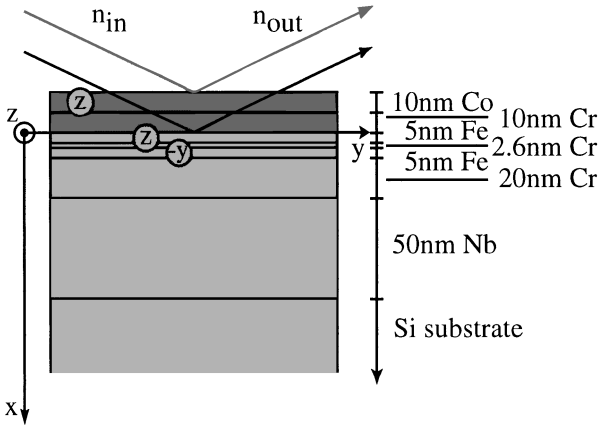

FIG. 1. Experimental arrangement for measuring the complex reflection matrix. The reference double layer is shaded dark. The symbols in circles indicate the direction of magnetization.

the polarization measurements to obtain $\mathcal{G}^{\mathrm{tot}}$. The upper index "tot" refers to the arrangement consisting of the sample and the reference layer. The reference layer is chosen here as a double layer built up of a magnetized film and a nonmagnetic spacer.

The reflection matrix $\mathcal{R}^{\mathrm{tot}}$ is related to the unknown reflection matrix of the sample $\mathcal{R}^{\mathrm{S}}$ by

$$\begin{aligned} \mathcal{R}^{\mathrm{tot}} &= \mathcal{R}^{\mathrm{ref}} + \mathcal{E}, \\ \mathcal{E} &= \mathcal{T}^{\mathrm{ref}}\mathcal{R}^{\mathrm{S}}[1 - \mathcal{R}_{\mathrm{R}}^{\mathrm{ref}}\mathcal{R}^{\mathrm{S}}]^{-1}\mathcal{T}^{\mathrm{ref}}. \end{aligned} \tag{11}$$

The reference layer is known and so are its transmission matrix $\mathcal{T}^{\mathrm{ref}}$ and its reflection matrices $\mathcal{R}^{\mathrm{ref}}$ and $\mathcal{R}_{\mathrm{R}}^{\mathrm{ref}}$. (The lower index "R" characterizes the reflection matrix for a beam impinging from the right; reflection matrices without the lower index correspond to the standard situation of an incident beam from the left.) The reflection matrix of the sample is given by $\mathcal{R}^{\mathrm{S}}$. Equation (11) is a generalization of Eq. (11) of Ref. [6] except for a shift of origin.

Using the factorization (9) for $\mathcal{R}^{\mathrm{S}}$ and $\mathcal{R}^{\mathrm{tot}}$ we must determine R_{++}^{S} from the four equations implied by the matrix relations (11). Elimination of $\mathcal{R}_{++}^{\mathrm{tot}}$ in (11) yields the three equations ($ij = +-, -+, --$),

$$E_{ij} = G_{ij}^{\mathrm{tot}}[R_{++}^{\mathrm{ref}} + E_{++}] - R_{ij}^{\mathrm{ref}}, \tag{12}$$

which are each equally suitable for the extraction of R_{++}^{S}. With the definitions

$$\begin{aligned} \mathcal{A} &= \mathcal{R}_R^{\mathrm{ref}}\mathcal{G}^S, \qquad \mathcal{B} = \mathcal{T}^{\mathrm{ref}}\mathcal{G}^{\mathrm{S}}\mathcal{T}^{\mathrm{ref}}, \\ C &= -\mathcal{T}^{\mathrm{ref}}(\mathcal{R}_R^{\mathrm{ref}})^{-1}\mathcal{T}^{\mathrm{ref}}, \end{aligned} \tag{13}$$

we can cast the matrix $\mathcal{E}$ of (11) into the form

$$\mathcal{E} = \frac{R_{++}^{\mathrm{S}}[\mathcal{B} + R_{++}^{\mathrm{S}}C\det(\mathcal{A})]}{1 - R_{++}^{\mathrm{S}}\mathrm{Tr}(\mathcal{A}) + (R_{++}^{\mathrm{S}})^2\det(\mathcal{A})}. \tag{14}$$

Substituting (14) in (12) yields three quadratic equations for $R^{S}_{++}(ij = +-, -+, --)$,

$$1 + \alpha_{ij}\mathcal{R}^{S}_{++} + \beta_{ij}(\mathcal{R}^{S}_{++})^2 = 0, \qquad (15)$$

with

$$\alpha_{ij} = \frac{G^{tot}_{ij}B_{++} - B_{ij}}{G^{tot}_{ij}R^{ref}_{++} - R^{ref}_{ij}} - \mathrm{Tr}(\mathcal{A}), \qquad (16)$$

$$\beta_{ij} = \det(\mathcal{A})\left[\frac{G^{tot}_{ij}C_{++} - C_{ij}}{G^{tot}_{ij}R^{ref}_{++} - R^{ref}_{ij}} + 1\right]. \qquad (17)$$

The three equations (15) are mathematically equivalent and have the same two roots. The stability of their solutions against measurement errors is different, however, and one chooses the most stable one for the analysis. The method works both above and below the critical value q_{cr} for total reflection. Of the two solutions R^{S}_{++} of (15) one selects the physical one by requiring that its phase ϕ^{S}_{++} tends towards the limit $-\pi$ as $q \to 0$. This selection by continuity requires polarization measurements over a range of momenta. From measurements at single q values one may select the physical solution by employing the criterion $\mathrm{Tr}(\rho) \leq 1$. This works for q values above the regime of total reflection, where it commonly turns out that only one solution satisfies this requirement. Unfortunately, we have no general proof of this.

To test the procedure we have applied it to a realistic example. Specifically, we consider a multilayer sample studied by Schreyer [10], consisting of five layers of different thickness (5 nm Fe, 2.6 nm Cr, 5 nm Fe, 20 nm Cr, and 50 nm Nb) mounted on a Si substrate. For the reference layer we take a double layer consisting of a 10 nm thick magnetized Co film and a 10 nm thick Cr film on top of the sample (see Fig. 1). The Cr layer is inserted to avoid a possible influence of the magnetized Co layer on the magnetization of the sample. The magnetization of the Co layer in the desired direction requires an external magnetic field.

We have chosen the z axis parallel to the direction of magnetization of the reference layer at the surface of the sample. The direction of propagation perpendicular to the surface defines the x axis. As to the magnetic field in the sample we assume the first Fe layer to be magnetized up to saturation in the z direction, and the second in the $-y$ direction.

Following the procedure outlined above, we have reconstructed the reflection matrix $\mathcal{R}^{S}$. We have considered incident beams polarized in the $\pm x$ and $+y$ directions as these seem to give the most stable solutions for configurations predominantly magnetized in the $+z$ direction. The reconstructed phases and the moduli squared of the matrix elements of $\mathcal{R}^{S}$ are displayed in Fig. 2 (solid curve). Only the physical solution is shown.

If we change the magnetization of the second Fe layer to the $+y$ direction, the phases of the nondiagonal elements of $\mathcal{R}^{S}$ are interchanged, but their moduli remain unaltered. This underlines the importance of the phase determination once again.

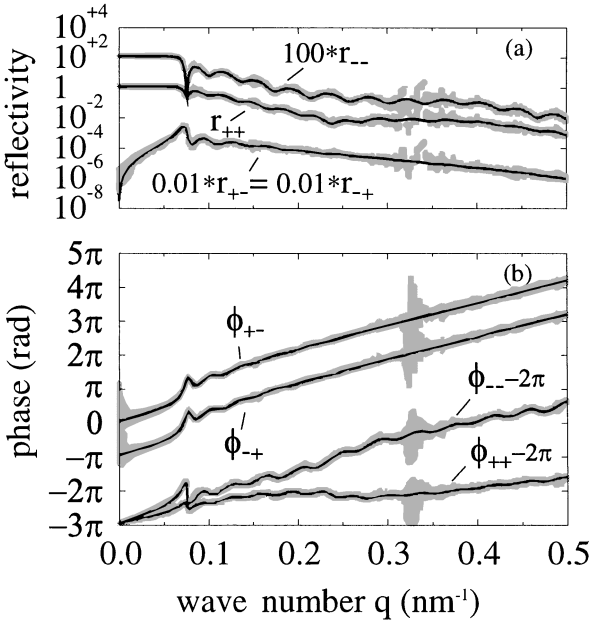

FIG. 2. Reconstruction of the reflection matrix R^{S} extracted from simulated polarization data for an arrangement with magnetic reference layer. The simulation includes the effect of absorption, surface roughness, and assumes a mean uncertainty of 0.01 of the polarization measurements. (a) Square of the moduli $r_{ij} = |R^{S}_{ij}|^2$, (b) phases ϕ_{ij}; $i, j = 1, 2$; $1 = +$, $2 = -$. The error bands correspond to a probability content of 68%, the solid curve represents the exact solution. For better visibility, r_{+-} and r_{-+} are divided by 100, r_{--} is multiplied by 100, and 2π is subtracted from ϕ_{++} and ϕ_{--}. For further details, see text.

So far we have used exact "data" and, therefore, the reconstruction must be perfect no matter which of the three equations (15) has been used in the procedure. We have simulated measurement errors by using input ensembles of normally distributed polarization values with half-width $\Delta P^{S}_{i}, \Delta P^{tot}_{i} = 0.01, i = x, y, z$. To include surface roughness we have randomly varied the positions of the interfaces within a width of 0.3 nm around the mean value. Figure 2 shows the reconstructed phases and the moduli squared of the matrix elements of $\mathcal{R}^{S}$; they are in fair agreement with the exact values. The error bands correspond to a probability content of 68% but they do not represent the standard deviation because the probability distribution is distorted. One finds isolated regions where the solution is rather unstable. This is due to the near vanishing of the discriminant $\frac{1}{4}\alpha^2_{ij} - \beta_{ij}$ in the quadratic equation (15).

We have repeated the previous calculation assuming that there is no magnetic field in the Co layer. The results are displayed in Fig. 3. The errors are slightly increased compared to Fig. 2, but still an excellent retrieval of the reflection matrix is achieved. These results demonstrate

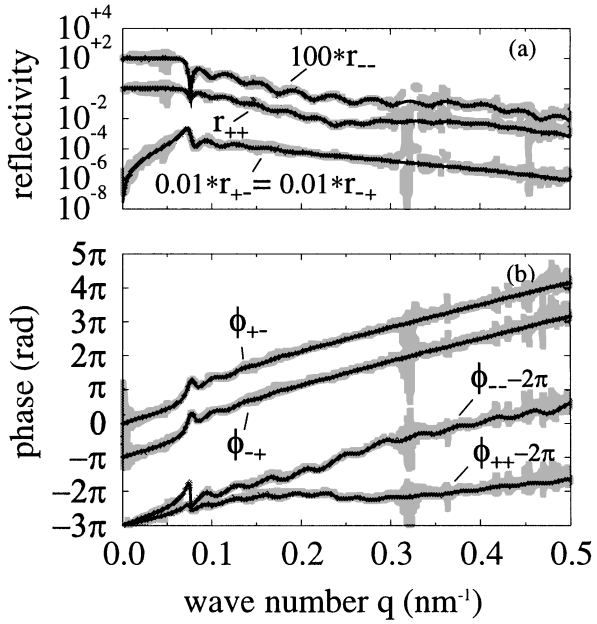

FIG. 3. Reconstruction of the reflection matrix R^S using a nonmagnetic reference layer. For more explanations cf. caption to Fig. 2.

that the use of a magnetized film within the reference layer may reduce the errors of the phase retrieval but is, in principle, required only for nonmagnetic samples.

In summary, the present method solves the phase problem of spin-polarized neutron reflection measurements for the first time. It is based on polarization measurements and makes use of a reference layer. In analogy to the single-channel method presented recently [6] one may use a magnetic reference layer but this is not required for samples with internal magnetic fields. The method also works in the region of total reflection as well as for absorptive layers. A stability test with simulated data results in a reasonably reliable reconstruction of the reflection matrix thus attesting to the viability of the whole method. Apart from the fact that, in general, measurements up to high q values (necessary for a good spatial resolution) may present a challenge for experiment, there are no specific problems expected for the experimental implementation of the method using modern neutron-optical devices. Limitations on the allowable thickness of the reference layer may be placed by the achievable monochromaticity and angular resolution of the reflectometer, which must suffice to resolve the so-called Kiessig oscillations associated with the reference layer. The novel method presented in this Letter is best suited to provide valuable insight into the magnetic structure of multilayers and superlattices because it provides the necessary information for an unambiguous reconstruction of the scattering-length density profile.

The authors thank Dr. A. Schreyer for many stimulating discussions. The work was supported by *Jubiläumsstiftung der Oesterreichischen Nationalbank*, Project No. 6212.

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