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Unique Analysis of Neutron Specular Reflection Measurements

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A recent proposal to determine the amplitude and phase of the reflection coefficient in neutron specular reflection experiments is generalized and discussed with regard to its applicability. It is shown that a phase determination is only possible if the magnetic reference layer is sufficiently thin, otherwise averaging due to the finite energy width of the beam will destroy the phase information.

KEYWORDS: Neutron Specular Reflection, Phase Problem, Surface Profiles

§.1 Introduction

The analysis of surface profiles by x-ray and neutron specular reflection is of scientific and technological interest.[1-4] Here the reflectivity, measured as a function of the wave number q of the incident beam, is used to determine the scattering-length density profile of the surface and thus the corresponding matter distribution.

Specular reflection can be formulated as a one-dimensional quantum-mechanical scattering problem (Fig. 1) characterized by the complex-valued reflection and transmission coefficients $R(q)$ and $T(q)$, respectively. The determination of the scattering-length density profile from the knowledge of $R(q)$ is a paradigm of an inverse scattering problem originally solved by Marchenko.[5] Specific solutions applicable to specular reflection experiments have been given by Kay[6] and later by Moses and de Ridder.[7]

Although the formal solution of the one-dimensional quantum-mechanical inverse scattering problem has been known for a long time, applications and numerical solutions have only recently been discussed.[8-13] The main reason for this appears to be the fact that knowledge of the full complex reflection coefficient (modulus and phase) is required in order to obtain unambiguous inversion results.[14-16] Standard reflection experiments, however, yield only the modulus of the reflection coefficients and consequently do not allow extraction of surface profiles uniquely without further information (usually a model simulation). This is the so-called *phase problem* known from diffraction analysis[17-20] which has so far prevented the model-independent analysis of reflection data.

Several papers dealing with the phase of $R(q)$ have appeared in the last years.[14-16,21-27] Focussing on neutron specular reflection, we mention three different proposals for an actual phase measurement: i) The reference layer method where one gains information on the reflection phase via the interference between the reflections from a known reference layer and the surface profile under investigation; this method is used in optics but can also be applied to neutron reflectivity measurements.[28,29] ii) The interferometric Lloyd's mirage technique which allows the determination of the reflection phase from the interference with a coherent reference beam.[29-31] iii) The measurement of the dwell time which is related to the reflection phase.[14] Recently, a very promising scheme using a magnetized reference layer for polarized neutrons has been put forward independently by two groups.[32,33] So far no proposal has been implemented experimentally.

In the present paper we discuss a general method of determining the reflection phase using polarized neutrons and a known external magnetic field. The superposition of the magnetic interaction of the neutron and the unknown neutron optical potential of the sample generates an interference term which contains information on the reflection phase. Measuring the reflectivity of polarized neutrons for different magnetic fields provides a set of correlated data which allows the separation of the phase of the reflection coefficient in the same way as in the reference layer method. In fact the recent proposal of Majkrzak et al.[32] and de Haan et al.[33] is a special case of the discussed scheme. For the successful application of the procedure it is essential to resolve the interference term. Therefore we have studied the influence of the finite energy width of the beam on the measured quantities.

§.2. Reflection coefficient for polarized neutrons

The specular reflection of low-energy neutrons is described by the one-dimensional Schrödinger equation[34]

$$\left[\frac{d^2}{dx^2}+q^2-V(x)\right]\varphi(x;q)=0 \tag{1}$$

in the coordinate x perpendicular to the planar surface of the sample, where q is the perpendicular momentum of the incident beam and $V(x)$ is the neutron optical potential profile. In the following we assume that the sample extends over the region $x > 0$ with its reflecting surface at $x = 0$, so that $V(x) = 0$ for $x < 0$. The reflection coefficient $R(q)$ is then given by

$$R(q)=\sqrt{r(q)}e^{i\phi(q)}=\frac{q+i\gamma(q)}{q-i\gamma(q)} \tag{2}$$

where $\gamma(q)$ is the logarithmic derivative of the Jost-type wave function $\psi(x;q)$ at $x = 0$.

If we place the experimental set-up in an external magnetic field $\vec{B}(x)$ the potential acting on a polarized neutron acquires an additional term

$$V(x)\rightarrow V(x)+U(x) \tag{3}$$

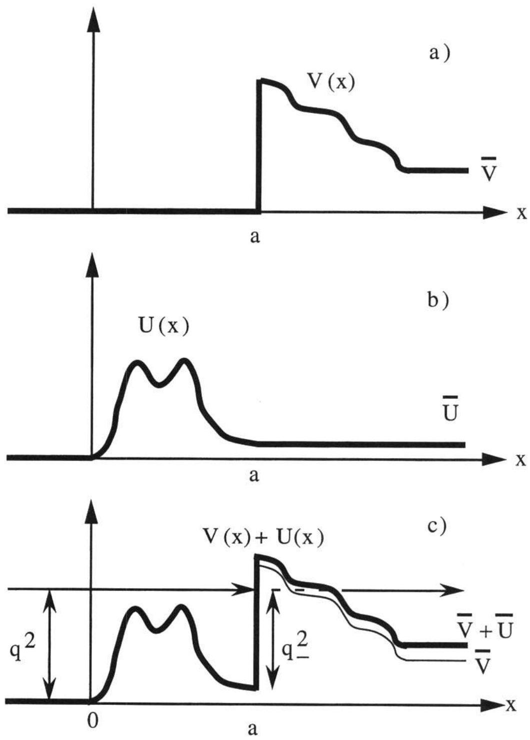

Fig.1 Sketch of the different interaction terms affecting the neutron specular reflection in the proposed set-up for σ_z=+1. a) Neutron optical potential; b) magnetic interaction $U(x)$; c) total interaction (heavy curve) and neutron optical potential (thin curve)

where

$$U(x) = -\frac{2m_n}{\hbar^2} g_n \mu_N \vec{\sigma} \cdot \vec{B}(x) \tag{4}$$

Here, μ_N is the nuclear magneton, g_n = -1.913 the gyromagnetic factor, and m_n the mass of the neutron.

The neutron optical potential $V(x)$ is independent of the external magnetic field. Thus specular reflection measurements under controlled variations of the magnetic field provide a set of reflectivity data which are related to each other, since they are generated by the same (but unknown) potential profile superposed with different known magnetic interactions.

In the following we use a coordinate system where the reflecting surface of the sample is shifted to $x = a$ so that the neutron optical potential vanishes for $x < a$ (see Fig. 1a). The keystone of the proposed procedure is the application of a magnetic field $\vec{B}(x) = B(x)\hat{z}$ perpendicular to the scattering plane which vanishes for $x < 0$ and is assumed homogeneous ($B(x) = \overline{B} > 0$) for $x > a$ leading to a constant magnetic interaction term

$$U(x) = \overline{U}\sigma_z, \ \overline{U} = \frac{2m_n}{\hbar^2}|g_n|\mu_N \overline{B} > 0 \tag{5}$$

within the sample. In the following we consider neutron beams polarized parallel and antiparallel to the magnetic field corresponding to σ_z = +1 and -1, respectively. The configuration is sketched in Fig. 1 for σ_z = +1.

The functional behaviour of the field $B(x)$ in the range $0 \le x \le a$ is arbitrary but known, and allows one to evaluate the (left, right) reflection and transmission coefficients $\rho^{\pm}_{L,R}(q)$ and $\tau^{\pm}_{L,R}(q)$ for the magnetic interaction alone. The indices (+) and (-) refer to the polarizations σ_z =+1 and -1, respectively. Knowing $\rho^{\pm}_{L,R}(q)$ and $\tau^{\pm}_{L,R}(q)$ it is possible to express the total reflection coefficient $R_{\pm}(q)$ in terms of the reflection coefficient $R(q)$ for the neutron optical potential $V(x)$ alone,

$$R_{\pm}(q) = \frac{\eta^{\pm}(q)R(q_{\mp})e^{2iaq_{\mp}} + \rho^{\pm}_L(q)}{1 - \rho^{\pm}_R(q)R(q_{\mp})e^{2iaq_{\mp}}} \tag{6}$$

with

$$\eta_{\pm}(q) = \tau^{\pm}_L(q)\tau^{\pm}_R(q) - \rho^{\pm}_L(q)\rho^{\pm}_R(q) \tag{7}$$

These relations are to be read separately for the upper and lower signs, where $q\pm$ is defined by

$$q_{\pm} = \sqrt{q^2 \pm \overline{U}} \tag{8}$$

The introduction of a homogeneous magnetic field for $x > a$ implies a modification of the beam energy within the sample, i.e. Eq. (6) relates $R_{\pm}(q)R(q_{\mp})$ (cf. also Fig.1c). The exponential $e^{2iaq_{\mp}}$ in Eq. (6) arises from the shift of the reflecting surface of the sample by the distance a (cf. Fig. 1), and $R(q)$ describes the reflection from the sample with its surface placed at the origin of the coordinate system.

In the special case of a steplike magnetic field ($U(x) = \pm\overline{U}$ for $x \ge 0$) the quantities $\rho^{\pm}_{L,R}$ go over into the Fresnel coefficients while $\eta^{\pm}$(q) becomes unity. Furthermore, Eq. (6) shows that for a magnetic field which sets in smoothly at $x = 0$ (so that the corresponding reflection becomes negligible, $\rho^{\pm}_{L,R} \ll 1$ and $\tau_{L,R} \approx 1$) the reflection coefficient $R_{\pm}(q)$ reduces to that of the (shifted) sample at the momentum $q_{\mp}$.

§.3 Determination of the complex reflection coefficient

In standard neutron specular reflection measurements only the reflectivity $r(q) = |R(q)|^2$ can be measured, while the reflection phase $\phi(q)$ remains undetermined. Here, we exploit the relationship between $R_{\pm}(q)$ and the reflection coefficient $R(q)$ for different magnetic interactions $U(x)$ to determine $\phi(q)$.

Taking the absolute square of Eq. (6) and replacing q with $q_{\pm}$, we obtain

$$\begin{aligned} &\left|1 - \rho^{\pm}_R(q_{\pm})R(q)e^{2iaq}\right|^2 r_{\pm}(q_{\pm}) \\ &= \left|\eta^{\pm}(q_{\pm})R(q)e^{2iaq} + \rho^{\pm}_L(q_{\pm})\right|^2 \end{aligned} \tag{9}$$

It is seen that the quantities $\rho^{\pm}_{L,R}$, $\eta^{\pm}$, and $r_{\pm}$ are always taken at the corresponding momenta $q_{\pm}$. We suppress these arguments in the following expressions.

For momenta in the range $q^2 < \overline{U}$ one must clearly use only the polarization $\sigma_z = +1$ associated with the argument

q_+, which remains real (while q_- becomes imaginary). Equation (9) can be rewritten as the equation of a circle in the complex R-plane,

$$|R(q) - z_\pm(q)|^2 = \varsigma_\pm^2(q) \qquad (10)$$

with the center at

$$z_\pm(q) = \frac{\rho_R^{\pm *} r_\pm + \eta^{\pm *} \rho_L^\pm}{r_\pm |\rho_R^\pm|^2 - |\eta_\pm|^2} e^{-2iaq} \qquad (11)$$

and the radius

$$\varsigma_\pm(q) = \frac{|\tau_L^\pm \tau_R^\pm|}{\left| r_\pm |\rho_R^\pm|^2 - |\eta_\pm|^2 \right|} \sqrt{r_\pm} \qquad (12)$$

Performing three experiments for each momentum q with the same sample but different magnetic fields, $B(x)$, and/or shifts of a, one obtains the necessary data to draw three circles in the complex R-plane with the complex number $R(q)$ as their intersection point. This method has already been proposed by de Haan et al.[33] for the special example of a homogeneous magnetic layer in front of the sample. It is essential for this method that the intersection point is well defined. Problems can arise at low momenta q. In fact de Haan et al.[33] observe a degeneracy of the three circles at low q -values thus limiting the possibility to extract the phase $\phi(q)$. However, as observed in Ref. 33 an extrapolation from q (0) = -π helps in this region.

§.4 Remarks on the applicability

In neutron specular reflection measurements we deal with neutron optical potentials in the range between $-20\cdot 10^{-4}$ nm^{-2} and $100\cdot 10^{-4}$ nm^{-2}. The neutron optical potential of most materials is repulsive and can therefore not sustain a bound state which would complicate the extraction of the potential from reflection data. In order to determine the reflection coefficient $R(q)$ by the procedure discussed above the magnetic interaction must not be negligible in comparison with typical strengths V of neutron optical potentials. For example for an Au-sample we have $V = 55.3\cdot 10^{-4}$ nm^{-2}; the equivalent field strength is $B \approx 3.4$ T. Usually much weaker magnetic fields ($B \approx 0.1$ - 0.3 T) will suffice to give rise to measureable effects. Such magnetic fields can easily be produced and present no obstacle for applications.

In order to extract the phase of the reflection coefficient $R(q)$ from Eqs. (10)-(12) via measurements of the reflectivity $r_\pm(q_\pm)$, the Kiessig oscillations in the latter must be resolved. These are associated with the factor e^{2iaq} appearing in the expression (6), whose absolute square yields the measured reflectivity $r_\pm(q_\pm)$. The Kiessig oscillations have a period π/a in the variable q, which must not be exceeded by the momentum resolution Δq of the neutron beam (determined by its angular resolution and monochromaticity),

$$\Delta q << \pi/a \qquad (13)$$

Therefore, the distance a must satisfy

$$a << \left(\pi/q\right)\left(\Delta q/q\right)^{-1} \qquad (14)$$

Depending on the type of selector used the monochromaticity part of the resolution $(\Delta q/q)$ lies between 0.1 and 10^{-4} or even lower, where the upper value refers to mechanical devices and the lower ones to crystal monochromators. For example, assuming a realistic beam monochromaticity $(\Delta q/q) = 10^{-2}$ only configurations with $a << 3\,\mu m$ at $q = 0.1\,nm^{-1}$ fulfil the condition (14). For larger distances a significant suppression of the phase information occurs.

If the Kiessig oscillations are averaged out the measured reflectivity $r_\pm(q)$ takes the form[35)]

$$\langle r_\pm(q_\pm)\rangle = |\rho_L^\pm|^2 + \frac{|\tau_L^\pm \tau_R^\pm|^2 |R(q)|^2}{1 - |\rho_R^\pm|^2 |R(q)|^2} \qquad (15)$$

Here, the phase information is completely lost.

§.5 Conclusions

In this paper we have generalized a recent proposal to determine the complete complex reflection coefficient for neutron specular reflection. This method exploits the superposition of the unknown neutron optical potential of the sample under investigation with a known magnetic interaction. One has to perform three neutron specular reflection measurements with polarized neutrons at a given q-value choosing different field configurations and/or displacements of the reflecting surface of the sample. The method seems most promising for sufficiently thin (<< 1 μm) magnetic layers directly on top of the reflecting surface of the sample. Such a configuration has been proposed by de Haan et al.[33)] One may also consider a set-up with a uniform magnetic field which extends over the entire positive x-axis. Shifting the sample by distances of the order of nanometers - which is possible today - one can scan a single Kiessig oscillation without resolving the full structure of $r_\pm(q)$. One still obtains the details of $R(q)$ by the intersection of at least three circles. With the full knowledge of $R(q)$ the neutron optical potential can be evaluated unambiguously.[11,36)]

We have seen that a main limitation of the method stems from the finite resolutions of the beam. However, the length scales involved are sufficiently large to promise interesting applications in various fields.

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