4 Multilayers

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Theme description:

In a wider sense the term *multilayer* (ML) describes a stack of thin (homogeneous) films of several materials.[1] The thickness of the individual layers reaches from a few Å up to the $\mu\mathrm{m}$-range; The layer materials can be insulators, metals, polymers, peptides or the like. This wide variety already tells that this is a wide field, reaching from biology (cell membranes are multilayers) over chemistry (sensors) to physics (optical devices, storage media). Here we refer to inorganic systems, used as reflective optical devices or as models for the investigation of magnetic or diffusion properties.

A. Properties of multilayers and reflectometry

A characteristic feature of multilayers is the strong anisotropy of their structure. In the direction perpendicular to the interfaces (z direction) the chemical composition changes and the characteristic length-scale is of that of 1 to several hundred unit cells. The deviations from a mean in-plane structure are due to formation of domains, non-smooth interfaces and strain. Within a layer the mean electron density or the mean scattering length density are constant. This means that the interfaces are of great importance for the properties of a ML.

An appropriate method for the investigation of interfaces are specular and non-specular reflectivity.[1, 2] The deviation of the refractive index of matter from 1 for cold neutrons (or for x-rays) is of the order of $\delta = 1 - n \approx 10^{-6}$. This means that most materials show a total external reflection up to $\theta_c \approx \sqrt{2\delta}$ of the order of $0.1°$, or in reciprocal space $q_c = 4\pi \sin\theta_c/\lambda \approx 0.01\,\text{Å}$. For $q > q_c$ the reflectivity R decays with $R(q_z) \propto (q_z - q_c)^{-4}$ for an ideal smooth (no lateral variation) and sharp (step-like change in the density) interface. Any deviation from a step-like density profile leads to an even faster decay. The specular reflectivity ($q = q_z$) contains information about the laterally averaged structure only! This has the consequence that a sharp, but rough interface leads to the same reflectivity curve as a diffuse smooth interface. The difference shows up in the off-specular ($q_y \neq 0$) region, where only the lateral structuring contributes.

If not only one interface is present the reflected waves interfere and R becomes modulated. This modulation is proportional to the Fourier transform of the structure factor squared of the scattering length density in z direction. If the layer sequence is periodic, $R(\theta)$ is comparable to the diffraction pattern of a 1-D crystal. Because of this similarity the maxima in R are called *Bragg peak*s, too. Periodicities of a length scale from 10 Å to 1000 Å can be resolved with cold neutrons.

B. Multilayers as optical devices

One application field for periodic MLs is the use as monochromators for neutrons (4.1) or x-rays (4.3). Unlike with crystals the lattice spacing can be adjusted to the needs and higher harmonics can be suppressed by adequately choosing the layer thicknesses or by modifying the interfaces (4.1). A third advantage is, that the lattice constant can be laterally graded, allowing for the production of focusing monochromators or the reduction of divergence (*Göbel mirror*).

If several multi-bilayers with different periodicity are deposited on top of each other several Bragg peaks occur. Thus the range of total external reflection can be enlarged to higher θ_c by an appropriate tuning of the layer thicknesses. Coatings of this kind are called *supermirror* (SM).[3, 4] They are used as coatings for neutron beam guides, or in optical devices where a wide wavelength band or a wide divergence is to be treated (4.4). Typical SMs consist of several hundred layers and reach $\theta_c^{SM} \approx 4 \times \theta_c^{Ni}$.

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[1] Sometimes *multi-bilayer systems* are called multilayers to distinguish them from *supermirrors*, which of cause also fit the definition.

C. Magnetic multilayers

If one of the layer materials is ferromagnetic, it is possible to design neutron optical devices with spin-dependent properties (4.4).

Another application field for multilayers is as model systems for the investigation of magnetic coupling (4.5, 4.6). *E.g.* bilayer systems consisting of materials of different magnetic properties (hard FM, soft FM, AFM, PM) are used to study coupling and exchange properties like exchange bias. Or trilayer systems, where hard and soft FMs are separated by a spacer are used to investigate the RKKY coupling. (4.5)

The project 4.6 is primarily concerned with the new physical phenomena that emerge when alternating layers of cuprate HTSC and manganite CMR materials or ferromagnetic SrRuO3 layers are brought in contact with each other. The goal is to understand the underlying physical interactions and ultimatively to tailore thin-film based materials with novel electronic and magnetic properties. These films have already been studied by magnetization, low energy muon spin rotation and optical techniques. It is expected to obtain valuable and complementary information by mapping out the field profile by neutron reflectivity.

D. Non-ideal multilayers

Real interfaces in a ML are neither sharp nor smooth. As mentioned in A, it is possible to deduce diffusion and roughness from reflectivity measurements. Interdiffusion between the adjacent layers causes a reduction of the Bragg peak height. This effect in turn can be used to determine interdiffusion with an effect to external treatment (*e.g.* heating, applying pressure, etc.). In particular, with the fact that scattering length densities vary significantly from isotope to isotope, it is possible to extract self-diffusen. An example for this is given in section 4.2.

E. Production

In our laboratory we have two sputtering plants which allow to deposit metallic or semiconducting material giving layer thicknesses down to 5 Å. The layers normally are amorphous or micro crystalline. To go to smaller thicknesses and to obtain crystalline films, other methods like MBE or CVD have to be used.

The advantages of sputtering the films are, that thick layers (up to several μm) are achievable, it is a fast process, and by an adequate choice of the geometry strong anisotropic in-plane stress can be induced in the films. This stress is the origin of an easy axis of magnetisation in ferromagnetic layers. This allows for the production of neutron beam polarisers (4.4).

In short the sputtering process goes as follows: In the sputtering chamber a glow discharge of Ar is induced. The cathode is formed (or covered) by the material to be sputtered. The Ar-cations are accelerated towards the cathode and hit neutral atoms out of it. These fast atoms hit the substrate and form the layer. The layer thickness can be adjusted via the power of the glow discharge, the velocity of the substrate underneath the cathode and the number of repetitions.

[1] T. P. RUSSELL. X-ray and neutron reflectivity for the investigation of polymers. *Material Science Reports* **5**, 171–271, 1990.

[2] V. HOLÝ, U. PIETSCH and T. BAUMBACH. *High-Resolution X-Ray Scattering from Thin Solid Films and Multilayers*. Springer Tracts in Modern Physics, Volume 149. Springer-Verlag Berlin Heidelberg New York, 1998.

[3] P. BÖNI, D. CLEMENS, MS. KUMAR and S. TIXIER. Challenges in the field of large-m supermirrors. *Physica B* **241**, 1060–1067, 1997.

[4] P. BÖNI. Supermirror-based beam devices. *Physica B Condensed Matter* **234-236**, 1038–43, 1997.