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Interference Coatings for Neutrons

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ABSTRACT

A review of multibilayer thin film devices for monochromating and/or polarizing neutrons with wavelengths of the order of 1\AA and longer is given.

The principle interactions of slow neutrons with condensed matter are nuclear and magnetic. For the present purpose, the neutron-nucleus interaction can be regarded as an isotropic point interaction characterized by a coherent scattering length b , the magnitude of which varies non-monotonically with different nuclei. Because the neutron is a spin $1/2$ particle and possesses a magnetic dipole moment of -1.913 nuclear magnetons, it interacts with those atomic electrons which give rise to a magnetic moment. This interaction can also be characterized by a scattering length p , although an angular form factor must, in general, be included due to the relatively large spatial extent of the interaction region. However, for scattering at the small angles of interest here, this form factor can be taken to be unity.

For scattering at small angles, then, the interaction of a neutron wave with matter can be described by a refractive index $n \equiv k'/k$ where k' and k are the magnitudes of the neutron wavevector inside the scattering medium and in vacuo, respectively. In free space $k = \sqrt{2mE}/\hbar$ where m is the mass of the neutron, E its energy, and \hbar is Planck's constant divided by 2π . Within a medium where the average interaction energy is $\langle U \rangle$, the magnitude of the neutron wavevector $k' = \sqrt{2m(E - \langle U \rangle)}/\hbar$. Thus,

$$n = \frac{k'}{k} = \sqrt{1 - \frac{\langle U \rangle}{E}} \quad (1)$$

For a saturated ferromagnetic material, which is of particular importance to polarizing applications of thin films, the refractive index can be expressed simply, in terms of scattering lengths, as

$$n = \sqrt{1 - \frac{\lambda^2 N}{\pi} (b \pm p)} \quad (2)$$

where λ is the neutron wavelength and N is the number of atoms/unit volume. The sign preceding the magnetic scattering length corresponds to one or the other of the two possible neutron spin eigenstates in an unpolarized beam. A ferromagnetic material is therefore birefringent. Although the absorption of neutrons is usually negligible, absorption can be treated in a conventional way by defining an imaginary part of the scattering length.

For most materials the nuclear coherent scattering length (or sum of nuclear and magnetic scattering lengths) is positive so that the refractive index is less than unity and total internal reflection from a mirror surface occurs. The critical angle θ_c corresponding to a material with a refractive index given by Eq. 2 is given by

$$\sin \theta_c = \lambda \sqrt{\frac{N}{\pi} (b \pm p)} \quad . \quad (3)$$

It turns out that one of the best materials for fabricating neutron mirrors for unpolarized neutron beams is unmagnetized Ni. In this case $\theta_c = 0.1^\circ / A \cdot \lambda(\text{\AA})$.

If a number of identical bilayers, each consisting of two thin films with different refractive indices, are superimposed, then diffraction of an incident neutron beam about an angle θ given by Bragg's formula $\lambda = 2d \sin \theta$ will occur where d is the thickness of the bilayer. This process can in fact be described by the kinematical theory for the diffraction of X-rays from crystals in the continuum limit if primary extinction is negligible and the diffraction peak is at an angle above the region of total internal reflection.¹ If extinction is not negligible, then dynamical theory must be used. These theories provide relatively simple analytic expressions which are

useful in understanding some of the general properties of multibilayers including reflectivity and polarizing efficiency (see, for example, Ref. 2). However, in order to account for mirror reflection and refraction effects, a method of calculation analogous to that encountered in the solution of thin film optical interference problems is appropriate which, in actuality, amounts to solving the Schrödinger equation for a neutron plane wave incident on and propagating through a layered but continuous medium.³ Boundary conditions are imposed at each interface and the reflection and transmission coefficients subsequently evaluated. Random as well as systematic variations in bilayer thickness can be readily incorporated.

The larger the difference in refractive indices of the two film materials, the greater the phase contrast and scattering power per bilayer becomes. For nonpolarizing multibilayer monochromators, one material is usually chosen with a large positive scattering length such as Fe and the other with a negative scattering length, for example Mn. It is also advantageous that the atomic density N of the material be high since the magnitude of the refractive index depends on the product of N and the scattering length.

It is possible, in addition, to polarize a neutron beam by diffraction from a multibilayer by choosing one film material, A, to be a saturated ferromagnet and the other some nonmagnetic material, B. The degree of polarization achieved then depends on the differences between $N_B b_B$ and $N_A(b_A \pm p_A)$. In the ideal case $N_B b_B = N_A(b_A - p_A)$ and one neutron spin state sees a uniform refractive index throughout the multibilayer and is not diffracted whereas the other spin state sees a modulated refractive index [$N_B b_B \neq N_A(b_A + p_A)$] and is, consequently, diffracted.

Multibilayers of Fe and Ge or one of a number of other nonmagnetic materials can have polarizing efficiencies of 98% or more.

Two general types of multilayer thin film devices for reflecting and polarizing neutron beams are presently in use. One device, already discussed above and hereon referred to simply as a "multilayer", consists of a number of bilayers with either a single bilayer thickness or distribution of thicknesses which give rise to a distinct diffraction peak. The use of multilayers for monochromating neutrons was first suggested by Schoenborn et al.⁴ The diffraction properties of such multilayers have been studied in some detail.^{1,5,6,7,8,9} As monochromators, multilayers have proven most useful at wavelengths of the order of 4\AA and longer. Polarizing multilayer monochromators were first investigated by Lynn et al.¹⁰ and Hamelin.¹¹ Subsequent development of multilayer polarizers is reported in References 7, 12, 13 and 14. Multilayer peak reflectivities approaching 0.9 and polarizing efficiencies greater than 98% have now been obtained for average bilayer thicknesses of the order of 50 to 100 \AA . Polarizing multilayers are especially valuable for polarized neutron scattering studies of condensed matter systems since high quality, conventional polarizers such as Heusler alloy single crystals are not widely available. Polarizing multilayers are also useful in avoiding adverse simultaneous or multiple scattering effects on polarizing efficiency which are sometimes encountered with mosaic crystal polarizers.¹³

A problem inherent to a multilayer composed of bilayers of a single thickness is that the angular acceptance for a given wavelength can be much narrower than the normal angular divergence of the incident beam. It has been shown, however, that the angular acceptance of a multilayer can be matched to

the beam divergence by deposition of an appropriate distribution of different bilayer thicknesses. Another limitation of multilayers is the relatively poor wavelength resolution that results for sufficiently short wavelengths, large bilayer thicknesses and divergent beams. Nonetheless, it is possible to significantly improve the wavelength resolution by using polarizing multilayers in conjunction with a wavelength - dependent spin flipping device. In fact, such an arrangement can actually be used to decouple wavelength resolution from the angular divergence of the beam.¹³

The other type of multiple thin film device has become known as a "supermirror" because it is composed of a particular sequence of bilayer thicknesses which in effect extends the region of total mirror reflection beyond the ordinary critical angle. This concept was introduced by Turchin¹⁵ and Mezei.¹⁶ Nonpolarizing supermirrors find application in neutron guide tubes. However, probably the most important application of supermirrors is as neutron polarizers. A substantial effort has been made in the development of supermirrors.^{17,18,6,19,20} At present, polarizing supermirrors have been constructed which extend the critical angle for one spin state to about 2.5 times that of an ordinary Ni mirror.²⁰

Finally, the microscopic chemical and magnetic structures of the thin films which compose a multibilayer can be interesting not only for their effects on the diffraction and polarizing properties of neutron optical devices but also in relation to fundamental problems pertaining to interfaces between two different materials.²¹ For example, in the case of FeGe multibilayers it is found, from higher angle X-ray diffraction measurements, that the Ge layers are amorphous whereas the Fe layers consist of microcrystallites with the ordinary bcc Fe structure but which are oriented

with a [110] direction normal to the plane of the substrate. More detailed investigation using polarized neutron scattering techniques reveals that there exists in addition a region of interdiffusion with an FeGe alloy structure in which the magnitudes of the Fe magnetic moments are significantly reduced.^{14,22} It is possible that such studies will be developed into a technique capable of providing important fundamental information on surface magnetic states.

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