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INVESTIGATION OF PERIODIC MULTILAYERS

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Боднарчук В. И. и др. E14-2009-127
Исследования многослойных периодических структур

В работе изучаются периодические тонкопленочные структуры, созданные по алгоритму, предложенному авторами. Методами нейтронной рефлектометрии определяются отражательные свойства полученных образцов, и проводится сравнение с теоретическими расчетами. Полученные результаты указывают на перспективность этих исследований.

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Investigation of Periodic Multilayers

Periodic multilayers of various periods were prepared according to an algorithm proposed by the authors. The reflectivity properties of these systems were investigated using neutron reflectometry. The obtained experimental results were compared with the theoretical expectations. In first approximation, the results proved the main features of the theoretical predictions. These promising results initiate further research of such systems.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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INTRODUCTION

Neutron supermirrors are nowadays in use in many neutron scattering experimental instruments. They consist of a system of multilayers usually composed as a set of bilayers of two materials possessing high and low optical potentials. The supermirrors increase the angular or wavelength range of total reflection comparing to mirrors consisting of a single material with high optical potential. If the single material gives total reflection for normal component k of the incident neutrons in the range $0 < k < k_c$, where k_c is the critical wave number for the given material, the supermirror can increase the interval up to nk_c. A multilayer system that gives reflectivity close to unity in the interval $0 < k < nk_c$ is called Mn mirror. In practice supermirrors are fabricated as M2, M3, etc., mirrors relatively to the k_c of nickel. However, there are several attempts to produce mirrors with higher n. The last record figure M6.7 was achieved by R. Maruyama et al. [1].

Last time all mirrors were prepared in aperiodical fashion, i.e., the thicknesses of layers in bilayers vary with the bilayer number. F. Mezei and P. A. Dagleish performed the first experimental study of such a supermirror in 1977 [2]. A modified algorithm for thickness variation was proposed by J. B. Hayter and H. A. Mook (HM) in 1989 [3]. According to this algorithm the change of thicknesses of neighboring bilayers is very small and does not match interatomic distance. It leads to creation of unavoidable roughnesses on layers interface. An alternative algorithm was proposed [4], according to which the supermirror is to be produced as a set of periodic stacks with some number N of identical bilayers. The variation of thicknesses of neighboring stacks in this case is larger compared to HM algorithm, which allows one to improve the quality of interfaces, and therefore the quality of supermirror as a whole.

The goal of the given work is to investigate how perfectly we can control the thickness and quality of periodic multilayer systems prepared by Mirrotron Ltd, Budapest. In other words, we want to see how the neutron reflectivity of produced systems matches theoretical expectations, how large is the diffuse scattering appearing due to the technological imperfectness aiming at trying to explain and control the values of the above properties.

Below we first present theoretical description of periodic multilayer systems, and calculate neutron reflectivities of periodic chains with N bilayers ($N = 2, 4, 8$). Further, we present results of measurements of reflectivities of fabricated multilayers and compare them to theoretical ones.

1. THEORETICAL DESCRIPTION OF PERIODIC CHAINS

A periodic chain consists of N bilayers every one of which contains two layers of different materials. In our case they were Ni and Ti. We call Ni with higher optical potential a «barrier», and accept the real part of its potential u_{b0}, which is equal to $2.45 \cdot 10^{-7}$ eV, for unity. It means that the other energies are defined in units of u_{b0}. So, the full Ni potential with imaginary part is $u_b = 1–i \cdot 3.47 \cdot 10^{-4}$. Then, we call Ti with lower potential a «well», and its optical potential in units of u_{b0} is $u_w = -0.203–i \cdot 1.325 \cdot 10^{-4}$.

It was decided to investigate periodic stacks, that give Bragg reflection at the point $k = 2$ given in units of the critical wave number $k_c = \sqrt{u_{b0}}$ of Ni. This point is the normal to the sample surface component of the incident neutrons wave vector. The point $k = 2$ has to be at the center of the Darwin table of the Bragg peak. Our main task is to find thicknesses of the two sublayers of a bilayer, to get Bragg peak (at $N \to \infty$) with maximal width of the Darwin table.

Reflection amplitude of a periodic potential with N symmetrical periods is given by the equation [4]:

$$R_N(k) = R_\infty \frac{1 - \exp(2iqNa)}{1 - R_\infty^2 \exp(2iqNa)}, \tag{1}$$

where

$$R_\infty = \frac{\sqrt{(1+r)^2 - t^2} - \sqrt{(1-r)^2 - t^2}}{\sqrt{(1+r)^2 - t^2} + \sqrt{(1-r)^2 - t^2}}, \tag{2}$$

$$e^{iqa} = \frac{\sqrt{(1+t)^2 - r^2} - \sqrt{(1-t)^2 - r^2}}{\sqrt{(1+t)^2 - r^2} + \sqrt{(1+t)^2 - r^2}} \tag{3}$$

and r, t are reflection and transmission amplitudes of a single period. In the case of a bilayer the potential of a period is not symmetric, as is shown in Fig. 1. Therefore, we have to take into account a direction of reflection and transmission. We denote $\overrightarrow{r}$ the reflection amplitude for the incident wave propagating to the right, and $\overleftarrow{r}$ for the incident wave propagating to the left. Then,

$$\overrightarrow{r} = r_b + t_b^2 \frac{r_w}{1 - r_b r_w}, \quad \overleftarrow{r} = r_w + t_w^2 \frac{r_b}{1 - r_b r_w}, \quad t = \frac{t_b t_w}{1 - r_b r_w}, \tag{4}$$

where

$$r_{b,w} = r_{0b,w} \frac{1 - \exp(2ik_{b,w} l_{b,w})}{1 - r_{0b,w}^2 \exp(2ik_{b,w} l_{b,w})},$$
$$t_{b,w} = \exp(ik_{b,w} l_{b,w}) \frac{1 - r_{0b,w}^2}{1 - r_{0b,w}^2 \exp(2ik_{b,w} l_{b,w})}, \tag{5}$$

$$r_{0b,w} = \frac{k - k_{b,w}}{k + k_{b,w}}, \quad k_{b,w} = \sqrt{k^2 - u_{b,w}}. \tag{6}$$

We see that the transmission amplitude t is symmetric, i.e., it is the same in both directions. Thus, we can also introduce the symmetrized reflection amplitude

$$r = \sqrt{\overrightarrow{r}\,\overleftarrow{r}}, \tag{7}$$

then, using the asymmetry property, Eq. (1) takes the form:

$$\overrightarrow{R}_N(k) = \overrightarrow{R}_\infty \frac{1 - \exp(2iqNa)}{1 - R_\infty^2 \exp(2iqNa)}, \tag{8}$$

where $\overrightarrow{R}_\infty$ and $\overrightarrow{R}_N$ inherit the asymmetry of r, i.e.,

$$\overrightarrow{R}_\infty = \frac{\overrightarrow{r}}{r} R_\infty, \quad \overrightarrow{R}_N = \frac{\overrightarrow{r}}{r} R_N \tag{9}$$

and R_N, R_∞ are given by (1), (2) with symmetrized amplitude (7) used for r.

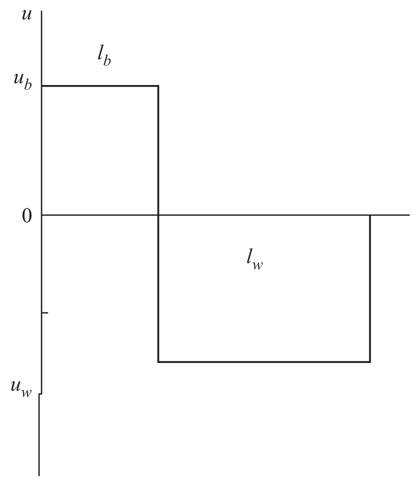

Fig. 1. A single element of a multilayered system is a bilayer composed of two different materials. The layer of one material has a high optical potential u_b and a width l_b. The layer of the other material has lower potential u_w and the width l_w

To find $l_{b,w}$ of the layers in the bilayer, which at $k = 2$ give the center of the widest possible Darwin table with $\left|\overrightarrow{R}_\infty\right| = 1$, we first neglect imaginary parts of the potentials $u_{b,w}$, and represent t in the form $t = |t| \exp(i\psi)$ with real phase ψ. Then, $r = \pm i|r| \exp(i\psi)$ with the same phase ψ, and Eq. (2) can be transformed to

$$R_\infty = \frac{\sqrt{\sin\psi(k) + |r(k)|} - \sqrt{\sin\psi(k) - |r(k)|}}{\sqrt{\sin\psi(k) + |r(k)|} + \sqrt{\sin\psi(k) - |r(k)|}}. \tag{10}$$

From it we see that the Bragg reflection takes place when $\sin^2\psi < |r|^2$, the center of the Darwin table is at $\sin\psi = 0$, and the larger is $|r|$, the wider is the Darwin table. Therefore, we must find the widths $l_{b,w}$ from two conditions:

$$\psi(l_b,\, l_w,\, k=2) = \pi,\ |r\,(2)| = \max\left(|r(l_b,\, l_w,\, k=2)|\right), \tag{11}$$

where we had shown dependence of ψ and $|r|$ on widths $l_{b,w}$. To have the conditional maximum t at the point $k = 2$ we are to require maximum at this point of the function:

$$F(l_b,\, l_w) = |r(l_b,\, l_w,\, k=2)| + \lambda[\psi(l_b,\, l_w,\, k=2) - \pi], \tag{12}$$

where λ is the Lagrange multiplier. The maximum is found from three equations:

$$\frac{d}{dl_b},\ \frac{d}{dl_w},\ \frac{d}{d\lambda}[|r| - \lambda(\psi - \pi)] = 0. \tag{13}$$

Solution of these equations gives

$$k_b l_b = k_w l_w = \frac{\pi}{2}. \tag{14}$$

Therefore, we must take

$$l_b = \frac{\pi}{2\sqrt{4-u_b}} = 0.907, \quad l_w = \frac{\pi}{2\sqrt{4-u_w}} = 0.766. \tag{15}$$

The unit of length corresponds to $k_c = 1$ (Ni), therefore it is $\lambda_c/2\_ = 92$ Å. So $l_b = 83.4$ Å, and $l_w = 70.4$ Å. It was decided to ask preparation of 3 samples with 2, 4 and 8 bilayers with thicknesses: Ni $l_b = 84$ Å, and Ti $l_w = 70$ Å.

2. MEASUREMENT AND PROCESSING OF DATA

The measurements on the five samples consisting of 1, 2, 4 and 8 Ni/Ti bilayers and pure substrate (floatglass with boron addition) as well were carried out at the neutron reflectometer REF in Budapest Neutron Centre (BNC). The scheme of the experiment is shown in Fig. 2. In this experiment the incident beam was monochromatized by crystal of pyrolytic graphite providing neutrons of wavelength equal to 4.28 Å. The sample mirror could be rotated to change the grazing angle θ of the incident neutrons. A position-sensitive detector was able to register the scattered neutrons over wide enough angular range for percepting all the neutrons entering the foreseen range of θ.

In Fig. 3 are shown the results of fitting experimental data for all samples with the formula:

$$R(k) = \int\limits_{k-\delta}^{k+\delta} \left|\vec{R}_{Ns}(p)\right|^2 \frac{dp}{2\delta} + n_b, \tag{16}$$

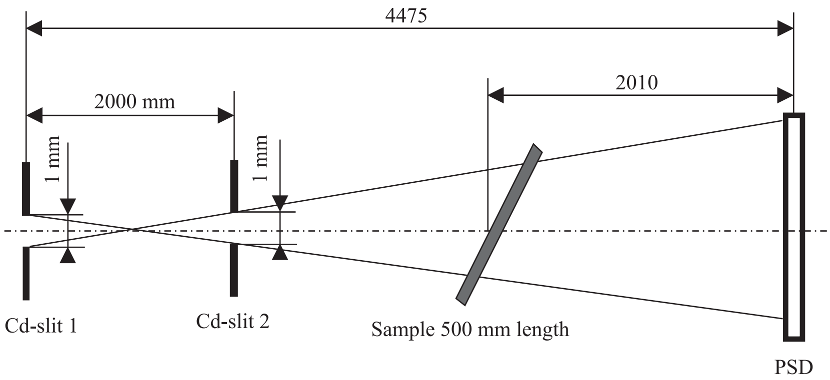

Fig. 2. The scheme of the neutron reflectometer REF in BNC. The sample mirror can be rotated around an axis perpendicular to the plane of the Fig. The position-sensitive detector is stationary and sufficiently large to register all the reflected neutrons, when grazing angle is sufficiently small according to experimental requirements. Collimation angle was 0.5 mrad and the neutron beam was monochromatic with wavelength 4.28 Å

Fitted values of real parts of potentials u' for Ni (b-barrier), Ti (w-well) and substrate (s), their thickness l, resolution δ, background n_b and χ^2 for periodic chains with $N = 2, 4, 8$ bilayers. Imaginary parts u'' of the potentials were fixed. The results are given in dimensionless units. The unit of energy is equal to real part of Ni optical potential $u'_{\mathrm{Ni}} = 0.245$ μeV, and unit of length is the reduced critical wavelength $\lambda_c/2\pi = 92$ Å for Ni

N	u'_b	u'_w	u'_s	u_b''	u_w''	u_s''	l_b	l_w	δ	n_b	χ^2
2	0.964	–0.258	0.452	0.00014	0.00012	0.0001	1.121	0.59	0.036	0.003	25
4	0.934	–0.388	0.446	0.00014	0.00012	0.0001	1.182	0.525	0.033	0.003	114
8	0.972	–0.349	0.408	0.00014	0.00012	0.0001	1.13	0.579	0.036	0.003	151

where δ is the parameter of the wave vector resolution and

$$\overrightarrow{R}_{Ns}(p) = \overrightarrow{R}_N(p) + T_N^2(p)\frac{r_{s0}(p)}{1-\overleftarrow{R}_N(p)r_{s0}(p)} \tag{17}$$

is the reflection amplitude from a periodic chain of N periods evaporated over a semi-infinite substrate, and the substrate reflection amplitude is

$$r_{s0} = \frac{k-k_s}{k+k_s}, \quad k_s = \sqrt{k^2 - u_s}. \tag{18}$$

Equation (16) takes into account the final resolution of the installation, scheme of which is shown in Fig. 2, and possible existence of background, n_b, in the system. The resolution δ and background n_b were two fitting parameters. Besides them we

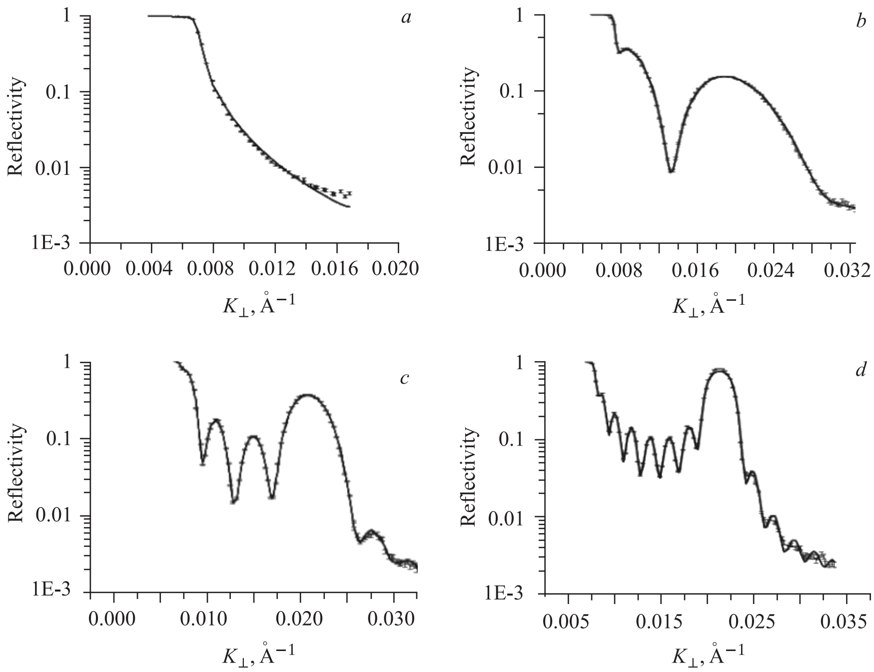

Fig. 3. The results of the fitting of the reflectivity data for periodic chains of bilayers evaporated on a boronised floatglass substrate. The fitting function is given by Eq. (16). From fitting of measured data to theoretical curves we could find the parameters of samples and resolution of reflectometer. Variation of parameters of samples shows to what extent the technological condition varies from sample to sample. In the figure: *a*) pure substrate (boronised floatglass); *b*) 2-bilayers film; *c*) 4-bilayers film; *d*) 8-bilayers film. There is good correspondence between experimental curves and calculation confirms the theoretical model. Some discrepancies at the right ends are caused by specific features of measurements

took as fitting parameters the real parts of all the potentials u_b, u_w, the potential of substrate u_s in terms of that of Ni, and thicknesses of Ni and Ti layers in units of $\lambda_c/2$ of Ni. Imaginary parts of the potentials were put to table ones: $u_b'' = 0.00014$, $u_w'' = 0.00012$, and $u_s'' = 0.0001$. The results for fitted parameters are presented in the first three lines of the Table. The last column of the table shows χ^2 of the fitting. The pictures in Fig. 3 show fit of all the samples, however the thicknesses of the Ni layers are more than 20% higher and the thicknesses of the Ti layers are more than 20% lower than in the project. The Ni potential was found to be slightly lower than is expected, which can be explained by presence of some oxygen or nitrogen impurities. The substrate potential $u_s \approx 0.4$ is too high comparing to pure silicon glass but for boron glass it is quite reasonable. The resolution $\delta \approx 3.5\%$ and background $n_b = 0.003$ are also quite acceptable. The worst was the value of χ^2. It is especially high in the case of the 8-periods sample.

Despite of high values of χ^2 the compliance of the theoretical model with the experimental curves is evident. The reasons of so high values of χ^2 are most probably caused by presence of the off-specular scattering that was added to the specular reflected neutrons. Due to some technical problems as a matter of fact we used the PSD detector as a single detector integrating the intensity over all surface of the detector. So at high values of $K_\perp$ when the reflectivity is about 10^{-3} the off-specular admixture distorts the experimental curve from its pure specular behaviour. Nevertheless, the first result gives us the confidence that this study has to be continued with opportunity of the separate measuring of off-specular and specular reflection.

CONCLUSION

Cooperation of theoreticians and experimentalists in research of multilayer systems is found to be very fruitful. We see that technology of preparation of such systems by Mirrotron Ltd, Budapest, is rather high quality, but it can be further improved taking into account the results of the analysis of surface imperfection and their correlation with parameters of producing systems. For that further analysis has to be carried out doing new experiments aimed at investigation of diffuse scattering and angular distribution of reflected neutron with better angular resolution.

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