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NEUTRON LOCALIZATION LIMIT, or FILM THICKNESS MEASUREMENTS BY NEUTRON REFLECTION METHOD

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Предел локализации нейтрона,
или Измерения толщины пленки методом отражения нейтронов

Обсуждается предельная чувствительность метода отражения нейтронов для измерения толщины тонких неоднородных слоев вещества на поверхности твердого тела. Показано, что чувствительность высокая и соответствует монослою твердого тела (т.е. около 0,5 нм), что находится на современном уровне умеренных требований к приборам. Введено понятие предела локализации нейтрона.

Работа выполнена в Лаборатории нейтронной физики им.И.М.Франка ОИЯИ.

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Kilany M., Chernenko L.P. E18-97-341
Neutron Localization Limit,
or Film Thickness Measurements by Neutron Reflection Method

The limit sensitivity of a neutron reflection method for thickness measurements of nonuniform thin layers of matter on the surface of solids is discussed. It is shown that the sensitivity is high and corresponds to the monolayer of a solid (i.e. about 0.5 nm), and is at a modern level of moderate requirements for tools. The concept of neutron localization limit is introduced.

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

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1. Introduction

It was shown [1] that the tunneling of polarized neutron wave-particles offers new possibilities for the investigation of surface diamagnetism in superconductors. As an example, a thin film Fabry-Perot resonator of natural vanadium sandwiched between two isotopical vanadium 50 thin films in the Meissner state, under total reflection, was considered. That method allowed the possibility of measuring strong magnetic penetration depths up to 4500 nm. The related sensitivity to penetration depth ratio and the external magnetic field was of the order of 10^{-6} and 1.2 Oe respectively.

But in [1], some distortion in flipping ratios - arising from nonuniformity of the thickness layer of multilayer structure - was not discussed.

Here, it is worthwhile to touch the sensitivity of a neutron reflection method for thickness measurements of nonuniform thin layers of matter on the surface of solids. Niobium thin film (about 80 nm thick) on a transparent substrate, will be considered as an example. The transparent substrate is chosen for neglecting the effect of reflection shadow.

Experimentally, it was already proved for the first time [2], that the sensitivity of neutron reflectometery method is rather high for nonuniformity in Nb thickness. Moreover, in view of the geometric arrangement of the sputtering apparatus, it was seen that a thin film of 28×50 mm^2 (sputtered on a silicon plate) had a graded thickness with an average value of 255 (+/- 15) nm. But the level of sensitivity of V-shaped profile for thickness measurements remains to be clarified.

2. Theoretical Background

Discreet media are commonly described by the neutron-classical potential interaction as

$$U = 4\pi \frac{\hbar^2}{2m} \bar{b} \rho \qquad \text{(i)}$$

where m is the neutron mass, $\bar{b}$ is the scattering length density, and ρ is the number of nuclei per unit volume.

The neutron reflection coefficient $R(k)$ as a function of wave-vector k, can be deduced by solving a one-dimensional quantum mechanical equation. Such approach to obtain the reflection coefficient is very efficient and has a great practical validity. But we will consider more realistic model of media concerning the reflection coefficient due to low energy neutron, reflected from the surface boundary of that model which contains N sequence of nuclear planes. Hence, equation (1) could be replaced by a singular one dimensional Fermi quasi-potential equation as

$$U(x) = \frac{\hbar^2}{2m} \sum_{n=1}^{N} \alpha_n \delta(x - x_n) \qquad (2)$$

where α_n is the average of nuclear scattering density in n-plane, x_n is the coordinate of that plane, and $\delta(x)$ is Dirac function. Schrodinger equation for such quasi-potential is equivalent to N recurrent equation [3] as

$$\psi_{n+1}'(x_n) - \psi_n'(x_n) = \alpha_n \psi_n(x_n), \qquad n = 1,2,\ldots,N$$

where N is the total number of planes. The continuity condition for neutron wave-function in the x_n point, creates an additional N equations as

$$\psi_{n+1}(x_n) - \psi_n(x_n) = 0, \qquad n = 1,2,\ldots,N$$

Let us put the neutron wave-function in a form of plane wave as

$$\psi_1(x) = A_1^+(k) e^{ikx} + A_1^-(k) e^{-ikx}, \qquad x < x_1$$

$$\psi_n(x) = A_n^+(k) e^{ikx} + A_n^-(k) e^{-ikx}, \qquad x_{n-1} < x < x_n, \qquad n = 1,2,\ldots,N$$

$$\psi_{N+1}(x) = A_{N+1}^+(k) e^{ikx} + A_{N+1}^-(k) e^{-ikx}, \qquad x > x_N$$

Taking the boundary condition into account,

$$A_{N+1}^-(k) = 0,$$

we can derive :

$$A_N^+ = \frac{\alpha_N}{2ik} A,$$

$$A_N^- = -\left(1 + \frac{\alpha_N}{2ik}\right) A e^{2ikx_N},$$

$$A_{n-1}^+ = -\frac{\alpha_n}{2ik} e^{-2ikx_{n-1}} A_n^- + \frac{2ik - \alpha_{n-1}}{2ik} A_n^+,$$

$$A_{n-1}^- = A_n^- + \left(A_n^+ - A_{n-1}^+\right) e^{2ikx_{n-1}} \qquad (3)$$

Equation (3) is simpler for programming than that described in ref. [3]. The reflection coefficient $R(k)$ can be expressed as

$$R(k) = \frac{\left|A_1^-(k)\right|^2}{\left|A_1^+(k)\right|^2}$$

in which the constant A is cancelled. A FORTRAN program is made specially, to carry out the calculations , using equation (3).

3. Results and Discussion

We discuss below our results for niobium thin film as follows:
For Nb $\alpha_1=\alpha_2=...\alpha_n=...\alpha_N=2.52\times10^{-5}$ nm^{-1} and its lattice constant $d = x_n - x_{n-1} = 0.4191$ nm. Let us take N=200.

Calculations were done taking into account the finite resolution of neutron reflectometer in moderate level of about 2.5%. For this, the convolution of reflection coefficient with a Gaussian-like resolution function was carried out. Fig. (1) shows the reflection coefficient of thermal neutrons in wave-vector range from 10^{-4} to 3×10^{-3} nm^{-1}.

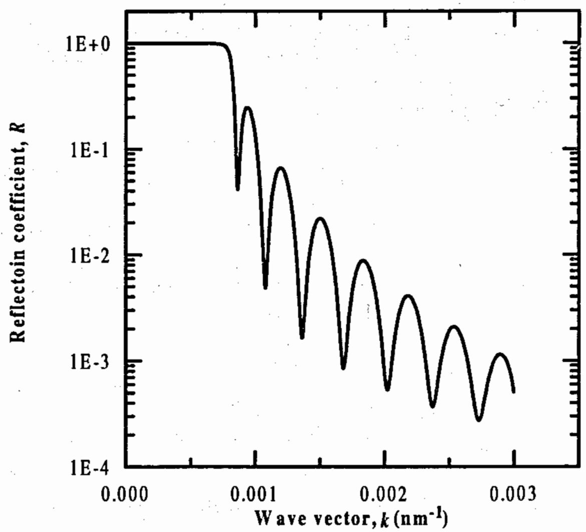

Fig. 1 Reflection coefficient R for Nb film of ~80 nm thick.

It is clearly seen that the total reflection region (R=1) exists in position where $k_o = k < 7\times10^{-4}$ nm^{-1} while interferences take place at values of $k > k_o$. This proves the validity of our program for reflectivity calculations, as well as the determination of its dynamical range which is experimentally measurable.

Let us consider two cases as follows: when N_1=200 and N_2=201, corresponding to R_1 and R_2 respectively. We take it because the magnetic penetration depth of Nb is of the same order of magnitude as thickness of such film and some interesting phenomena can take place at this thickness. But it is out of the scope in our publication. Fig. (2) shows the ratio R_2 / R_1 as a function of wave-vector k in the same range as previous figure.

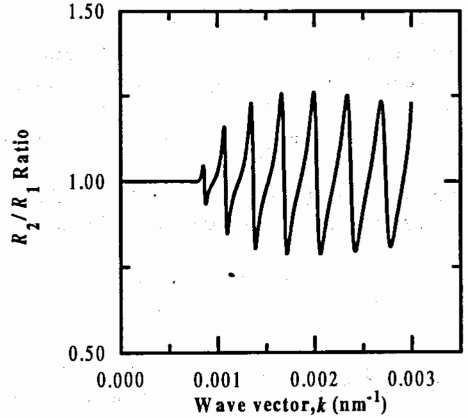

Fig. 2 Reflection coefficient ratio for N_1 = 200 and N_2=201 (R_1, R_2 respectively), according to a difference in film thickness of ~ 0.5%.

We will consider the case when 10^4 neutrons are reflected per each wave-vector value , as a simple experimental arrangement for Reflex installation [4], at IBR-2 Pulsed Reactor in Dubna. In this case , the Poisson error will be ~1% . From fig. (2) we can deduce that the sensitivity of neutron reflection measurement exceeds the level of ~1% , for a difference of thickness in one monolayer .

Let us consider a non-trivial case , when a thin Nb film is cooled or heated within 30 K. In such case , a lattice compression or expansion will be occurred respectively due to the thermal process , and the temperature deviation leads to a relative potential difference of ~0.001. We calculated the reflectivity R_1 and R_2 due to that potential difference . Fig. (3) shows the ratio R_1 / R_2 , which fluctuates nearly in the range of 1% , determining the limit of neutron localization for Nb film of ~80 nm thick.

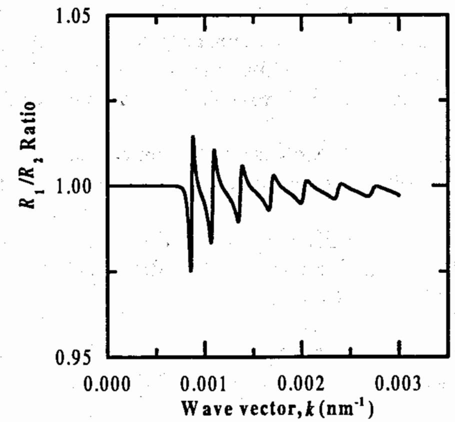

Fig. 3 Reflection coefficient ratio for two states of Nb film, at room temperature (R_1) and cooled 30K down (R_2).

We can say that there are no limit at all, if some differences in conditions are exist. Limitation occurs only once deviation appears during measurement. This limit is relative and varying according to the increase in sensitivity (by high resolution neutron reflectometry [1,5] and/or the increase in film thickness). A small factor of absorption and the surface roughness as a subject of real experiment could be neglected in these relative measurements, on neutron reflection.

Considering this preliminary calculations, one can show that there is a condition for increasing the sensitivity of reflection measurements. Further considerations could be appeared in later publication.

4. Conclusions

We considered the limit sensitivity of conventional neutron reflection method, for thickness measurements of nonuniform thin layers of niobium on transparent substrate. It is shown that the sensitivity is high for relative measurements, according to the monolayer of Nb (~0.5 nm), and lies in a modern level of moderate requirements of tools (neutrons, X-ray, electrons or ions). The concept of neutron localization limit is introduced.

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